MINISTERIO DE INDUSTRIA, COMERCIO Y TURISMO

CENTRO DE INVESTIGACIONES ENERGETICAS
MEDIOAMBIENTALES Y TECNOLOGICAS
(C.I.E.M.A.T.)



Dear Dr. Pettengill,

I am sending you a preliminary report on the whole (26 years) Indalo project, that could be of help to both of our organisations un order to identifity and structure our common future work.

We consider october as good option for meeting in Washington. Early in september we hould contact Mr Bell to define both the visit and the meeting.

Sincerely,

Francisco Mingot Buades
DIRECTOR INSTITUTO MEDIO AMBIENTE

Do you have the latest draft + final DRAFT ?

CENTRO
DE INVESTIGACIONES
ENERGETICAS
MEDIOAMBIENTALES
Y TECNOLOGICAS

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Instituto de Medio Ambiente

CIEMAT/IMA/UGIA/M5A01/03/92

# SUMMARY REPORT ON THE 26 PALOMARES SURVEILLANCE PROGRAM

DRAFT. VERSION 0

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Madrid Julio 1992

## 1. INTRODUCTION

As a consequence of the accident ocurred on January 1966 in the area of Palomares (Spain), actions aiming the radiological protection of the population were stablished. In addition to decontamination operations and countermeasures, wich are not the objective of this paper, a Radiological Surveillance Program (I.P.) concerning population and enviroment was initiated inmediately after the accident ocurred. This program has been carried out, without interruption until now, in the frame of Otero-Hall Agreement (USA/SPAIN).

At a National level CIEMAT has been technical responsible for the desing and execution of work. The Spanish Consejo de Seguridad Nuclear, institution responsible for radiation protection aspects in the country, must be informed periodically by CIEMAT.

Jointly with the Surveillance Programme and taking advantages of a real scenario, experimental research activities on the field of Radioecology have been running. This associated research is thought to be essential in order to gain knowledge on the environmental behaviour of Plutonium and Americium, to improve estimations of individual doses to the people living and working there and in this way to keep suitably updated the Surveillance Program.

The present report summarizes the main activities carried out in relation with Indalo Project. Also the most importants results and conclusions obtained so far are presented. From these results, needs for further studies are identified.

Therefore it is not an extensive report containing all individuals data obtained and a complete description of activities and methodologies that would be out of the scope. However references supporting the work realized and the results reported are included.

Finally it is considered that this report could be a suitable basis for discussion in order to reach a future collaboration agreement.

## 2.1. FOREWORD

In accordance with the specifications laid down in the Otero-Hall Agreement, as from when the remedial actions for the area were finished, a start was made on the activities aimed at studying the direct consequences of the accident and of the effects which, in the future, might derive from it, as well as from the residual radioactive contamination in the area. It order to fulfil this purpose a SURVEILLANCE PROGRAMME was set up, having been developed by the Junta de Energía Nuclear (JEN) in its capacity as the government body technically able to carry out the appropiate surveillance tasks and which, since 1986 have been performed by the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT), body rataimine the technical capacities of the JEN and having been apointed to perform the surveillance programme.

Given the characteristics of the area's contaminating radionuclides, all of which are alpha emitters while some emit very low-energy photons, the risks of external irradiation for the inhabitants of the zone, which might derive from the residual contamination, is practically negligible. Irradiation for those persons who live in or cultivate the area could only derive from its being taken into the organism through inhalation or ingestion.

As a result of the above conclusion, the following main objectives were set up:

- To determine the magnitude of the risk of internal contamination for the inhabitants of the zone during the period immediately following the accident and the subsequent emergency phase.

- To assess at short, medium and long-term the risk of internal contamination for those people living in and around Palomares, for those who cultivate the contaminated land and for those who consume vegetable products grown in this area, as well as products from animals which have been given as fodder cereals and other vegetables grown in the area.

In order to attain these objectives, the wideranging SURVEILLANCE PROGRAMME was drawn up and has been in constant operation since 1 June 1966.

This surveillance programme is based both on performing direct contamination measurements on people, and on keeping track of the evolution of the contamination in the soil and in the means and products that might give rise to the contaminating radionuclides being intaked by the human beings.

As a result, the activities to be carried out are included under the following headings:

- Soil.
- Air.
- Vegetation.
- Animals.
- People.

## 2.2. SOIL

Figure 2.1 shows the area contaminated by the accident, approximately 226 hectares, and how the measured alpha contamination spread before the countermeasures decided upon were undertaken.

A gross estimate of the plutonium inventory deposited in the zone on the basis of the specified contamination values and of the surface area affected made it possible to deduce that, excluding the two areas close to where the two fractionated bombs had fallen and where the maximum contamination levels were to be found, a minimum of $2 \times 10^{11}$ Bq (~5Ci) would remain in the zone as residual contamination once the countermeasures decided upon had been applied. These countermeasures included eliminating all the bomb fragments and the removal of a 10-cm surface layer of soil in those areas with the greatest surface contamination.

In order to determine how the concentrations of activity were spread in the soil as a result of the countermeasures undertaken, and how they have evolved with the passing of

time, six plots were chosen as being representative of the situation in each of the three zones into wich the contaminated area was divided. The locations of the plots referred to as 2-1, 2-2, 5-1, 3-1 and 3-2 are specified in figure 2.1.

The plots are quadrangular in shape with sides measuring 50 metres, and the soil samples taken from each of them correspond to nine points situated on the diagonals and equidistant from each other. The samples were cylindrical with a diameter of 31 mm and a depth of 45 cm. Each sample was divided into five fractions representing depths of 0-5, 5-15, 15-25, 25-35 and 34-35 cm.

The analyses carried out with each of the fractions obtained, these amounting to a total of 2,160 samples, and which were periodically collected during the time elapsed, have given the average values of the plutonium and americium concentrations in the 45-cm soil layer in these plots. These values are summarized in tables 2.1 and 2.2.

Regardless of the soil samples in these plots, and as a result of the observation and conclusions arising out of the surveillance plan, soil sampling have been performed that were specially designed to determine the spread of the contamination outside the original 0 line, and in particular to estimate the residual contamination in the area close to impact 2 which had basically been considered as having a low level of residual contamination due to the surface layer of soil having been removed as a countermeasure.

This zone, known as 2-0, which covers approximately 6.5 Ha from the impact-2 zone to the 2-1 sample plot, showed levels of Pu-239 + Pu-240 and Am-241 contamination that were higher than anticipated when the countermeasures undertaken in 1966 were completed. This fact led to a radiometric study of the zone, on the basis of measuring the low-energy gamma radiation given off by the Am-241, with Harshaw Gamma Counter field equipment (Mod. 301) known as FIDLER. The measurements were taken according to 2-metre-long grids over an extension of approximately 3.5 Ha, the most contaminated, and according to 10-metre-long grids for the others. A three-dimensional illustration of the results of the measurements performed to determine the relative contamination existing in this area is given in figure 2.2.

On the basis of this radiometric plan, 37 points were chosen in the zone for the purpose of taking soil samples down to a depth of 45 cm. Five fractions were taken in each sample, in terms of depth, and for each of these francions a radiochemical method was used to determine the Pu-239 + Pu-240 concentration, while the Am-241 concentration was determined by low-energy photon spectrometry.

The results of the tests have made it possible to conclude that, in the zone known as 2-0, the characteristics of the contamination down to a depth of 45 cm are as follows:

- The concentrations of plutonium and americium in these zones are not distributed in a homogenous manner. In the valley area they are found to a depth of 45 cm, whereas in the hills the concentration below the first 5-cm layer is less than 1 or more orders of magnitude.

- The average concentration of Pu-239 + Pu-240 is around 82 kBq/kg, with a maximum value of 681 kBq/kg close to point of impact 2 and a minimum value of 0.28 kBq/kg at a point in the hills.

- The average concentration of Am-241 is around 13.4 kBq/kg, with a maximum value of 135 kBq/kg close to point of impact 2 and a minimum value of 0.10 kBq/kg.

A gross estimate of the plutonium content in this zone has made it possible to conclude that over an extension of approximately 6.5 hectares an activity of about $6 \times 10^{11}$ Bq ($\sim$ 17Ci) can be found. Now, this appraisal will be made with greater precision.

It should be borne in mind that zone 2-0, apart from being the most highly contaminated as a result of the accident, was chosen for the canning of all the heavily-contaminated soil and vegetation as well as being the site where part of the highly-contaminated vegetation was buried.

In orden to give an idea of the size of the surveillance programme carried out in relation to the soil between 1966 and 1991, table 2.3 shows the number of soil samples and

radiochemical analyses that have been made.

## 2.3 AIR

The programme aimed at determining the level of contamination in the area's atmosphere in order to establish the risk of inhalation for people living in and cultivating the area has been carried out on a continous basis ever since the countermeasures were concluded.

For this purpose sampling stations were put in place, as shown in figure 2.1, and were referred to as 2-1, 2-2 P and 3-2, according to where they were located in the three reference zones that had been established. All of them were started up in 1966.

Stations 2-2 and P have been taking air samples continously since 1966. Stations 3-1 and 2-1 finished sampling in October 1969 due to a number of problems regarding electricity supply. Sampling in station 2-1 began once more in 1984 on account of the interest prompted by its locations.

The air samples were taken until 1981 on cellulose filters with a diamter of 47 mm, placed at a height of 1.70 m. They were changed on a daily basis and corresponded to an average air volume of 90 cubic metres every 24 hours.

The radiochemical analyses and the alpha spectrometry measurements to determine the concentration of Pu-239 + Pu-240 were performed on samples corresponding to 10-day periods in each of the stations and which, as a result, corresponded to air volumes of approximately 900 cubic metres.

In January 1981 the sampling systems were replaced by others with a greater aspiration capacity which have made it possible to collect aerosols corresponding to 10,000 cubic metres per week in asbestos filters with a surface area of 500 square centimeters.

Radiochemical analyses have been performed since 1981 on each of these weekly samples. This has made it possible to reduce the detection limit of Pu-239 + Pu-240 in the

radiochemical-separation and measurement by alpha spectrometry combination.

All these air samples took in all the existing aerosol particles, and their direct correlation with the risk of inhalation implied an overestimate of the risk. For this reason, in April 1987, stations P and 2-2 were provided with a sampling system which, since then, has made it possible to collect on the asbestos filter particles of a size of under 10 $\mu$m and which constitute the really inhalable fraction.

Table 2.4 shows the numbers of air samples and analyses (radiochemical separation plus measurement by alpha spectrometry) performed up to December 1991, this giving and idea of the size of this air monitoring programme.

The average concentration values for Pu-239 + Pu-240 corresponding to each of the sampling stations during each of the years included in the period between 1966 and 1990 are specified in table 2.5.

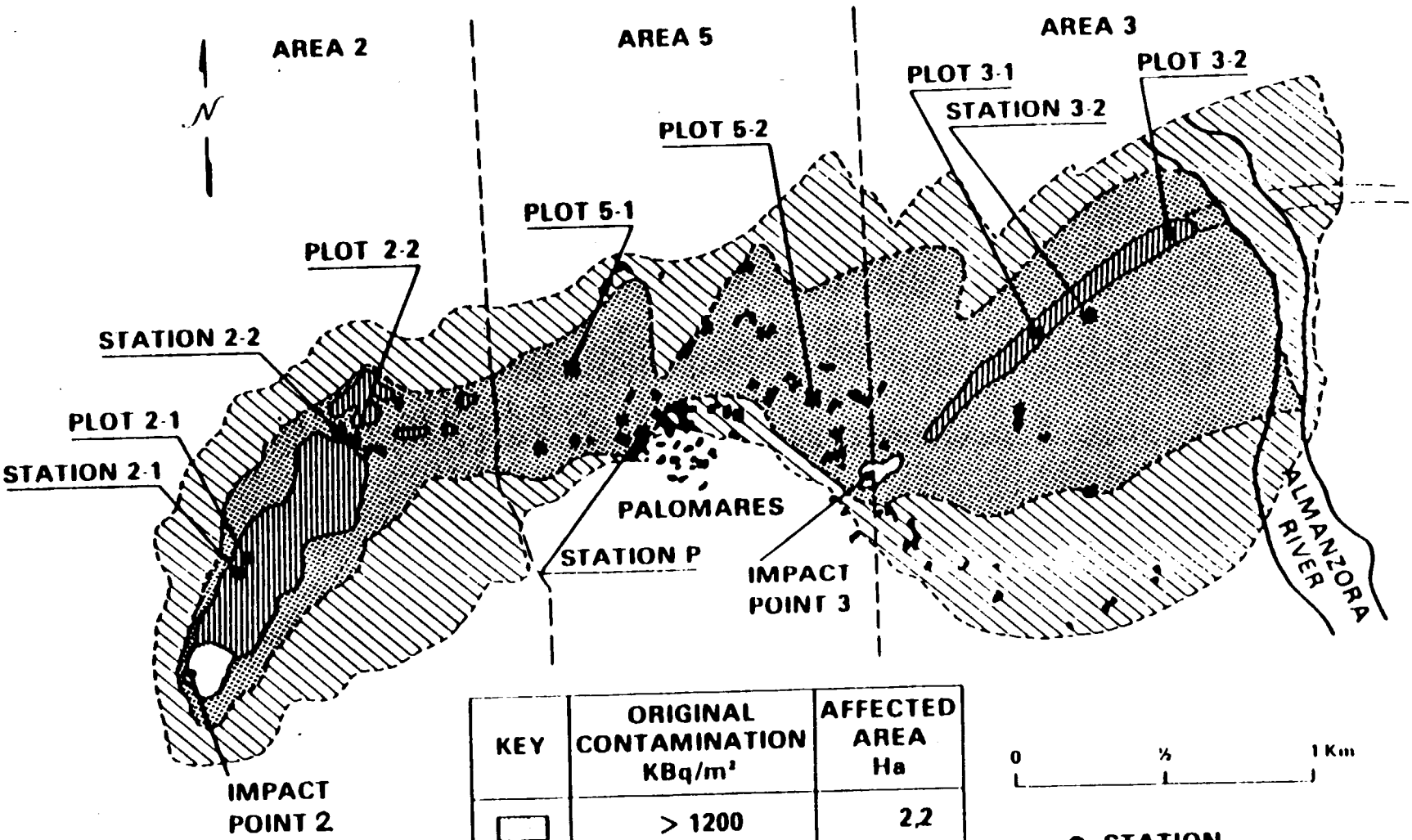
## 2.4. VEGETATION

The activities carried out under the Surveillance Programme with respect to vegetation have been aimed from the very beginning at ascertaining the level of plutonium contamination in both cultivated and wild plants growing in the area, specially as regards their edible parts. Since 1983 tests have also been carried out in order to determine the levels of americium contamination.

Until 1978 the plants were collected from the same plots chosen for studying soil contamination, in other words the six study plots shown in figure 1 with the reference numbers 2-1, 2-2, 5-1, 5-2, 3-1 and 3-2. Due to the fact that, for various reasons, these plots were not cultivated every year, it was decided in 1978 to extend the plant-sampling process to the areas close to these plots and which had similar levels of residual contamination. In recent years some crop-growing in the area has taken place in hothouses, especially for tomatoes, cucumbers and peppers, and so it was decided also to take samples of plants grown in hothouses in order to ascertain the differences that might result from enclosed cultivation systems.

Fig 1

# PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS AND LOCATION OF SAMPLING STATIONS




| KEY | ORIGINAL CONTAMINATION KBq/m² | AFFECTED AREA Ha |
|---|---|---|
| | > 1200 | 2,2 |
| | 120-1200 | 17 |
| | 12-120 | 87 |
| | < 12 | 120 |

0 ½ 1 Km

● STATION
■ PLOT

So as to give an idea of the size of the plant monitoring programme, table 2.6 shows the number of samples taken between 1966 and 1991 as well as the radiochemical analyses that were performed. In general, each sample corresponds to a wet weight of at least 3 kilos.

Table 2.7 shows the concentration values of Pu-239 + Pu-240 in the various plants grown in the area for the period 1966-1986 and which are typical Mediterranean products. The results obtained in the same crops for the period 1986-1991 does not modified in a significative way the average values.

Studying the results of the plutonium concentration in the plant samples, with most of the contamination affecting the parts with the greatest surface area or with a rough surface, and with a high degree of inhomogeneity among plants growing in the same area, led us to conclude that the contamination process is not due, in the vast majority of cases, to absortion through the roots. External contamination is predominant and, as a result, resuspension processes are an important factor.

The wild vegetation in plot 2-1, which is the nearest to point of impact 2 and which was decontaminated with manual tools on account of the nature of the soil, as well as the vegetation collected around this plot and in the S0 called 2-0 zone, has shown plutonium contamination in practically all the samples that have been taken. The highest concentrations have been found in esparto grass. The average, maximum and minimum values corresponding to the 114 esparto samples that have been taken are as follows:

| | | |
|---|---|---|
| Average value | = | 575 Bq/kg |
| Maximum value | = | 26 kBq/kg |
| Minimum value | = | 0.04 Bq/kg |

## 2.5. ANIMALS

The study of contamination in animals as part of the surveillance programme has been the least developed to date. Nevertheless, samples have been taken and radiochemical analyses performed for the purpose of ascertaining the contamination risk for certain animals native to the are and for animals that graze in the area or feed on plants growing wild or

cultivated in the area.

With this aim in mind tests have been performed to determine the level of contamination in snails from the various zones, in milk from goats that have grazed over the whole area, in the tissues of some of these goats and in henns and chickens that ranged in semi-freedom on a farm located in zone 2-2.

The sets of samples and radiochemical analyses corresponding to the monitoring of animals are given in table 2.8.

## 2.6. PEOPLE

The surveillance programme on human beings was set up for the purpose of providing a direct estimate of the level of internal plutonium contamination affecting the inhabitants of Palomares as a result of their subsequently remaining and carrying on their activities in the area.

This undertaking was planned for a long period of time, equivalent to the average life span of the people living in the area, and is aimed at monitoring all the inhabitants of Palomares. Internal contamination measurements and clinical/medical examinations are performed on those people who undergo annual testing.

Testing on people was begun in 1967 and, since 1975, has been performed annually without a break on approximately 150 people a year.

Measuring internal contamination in human beings used to be done by means of direct plutonium contamination measurements on the lungs, using a specific lung counter, and by determining the excretion of plutonium in the urine. Owing to the fact that the lung counter failed to detect -in any person- an amount of plutonium higher than the detection limit of 814 Bq, these measurements have not been carried out on a routine basis since 1988, and are only performed on people being tested for the first time. Between 1966 and 1990 a total of 1,190 measurements have been performed on 800 persons.

Since 1985 everyone has been tested for americium-241 excreted in the urine.

Table 2.9 gives the figures showing the size of the surveillance programme on human beings, both as regards the people as a whole and on an individual basis, for the period 1966-1989.

The results of the excretion of Pu-239 + Pu-240 in the urine, and which were obtained by means of the radiochemical analyses performed for th period 1966-1991, are set out in table 2.10.

Table 2.1.- PLUTONIUM CONCENTRATION IN SOILS OF THE SELECTED PLOTS.

| PLOT | Pu-239 + Pu-240 kBq/kg | | |
|---|---|---|---|
| | AVERAGE | MAXIMUM | MINIMUM |
| 2-1 | 0,44 | 1,60 | 0,03 |
| 2-2 | 2,06 | 3,31 | 0,80 |
| 3-1 | 1,10 | 2,00 | 0,05 |
| 3-2 | 1,79 | 5,70 | 0,23 |
| 5-1 | 0,13 | 0,30 | 0,02 |
| 5-2 | 0,29 | 0,99 | 0,01 |
| zone 2-0 | 82,00 | 681,00 | 0,28 |

TABLE 2.2.- AMERICIUM CONCENTRATION IN SOILS OF THE SELECTED PLOTS.

| PLOT | Am-241 kBq/kg | | |
|---|---|---|---|
| | AVERAGE | MAXIMUM | MINIMUM |
| 2-1 | 0,07 | 0,21 | 0,01 |
| 2-2 | 0,55 | 0,64 | 0,40 |
| 3-1 | 0,24 | 0,53 | 0,14 |
| 3-2 | 0,49 | 0,87 | 0,16 |
| 5-1 | 0,04 | 0,09 | 0,01 |
| 5-2 | 0,09 | 0,13 | 0,06 |
| zone 2-0 | 13,40 | 135,00 | 0,10 |

TABLE 2.3.- QUANTIFICATION OF THE SOILS SURVEILLANCE PROGRAM IN THE PERIOD 1966-1991.

| LOCATION | SAMPLES | ANALYSES | | |
|---|---|---|---|---|
| | | $\alpha$ Activity | Pu-239+Pu-234 | Am-241 |
| Surface | 994 | 126 | 885 | 351 |
| Depth | 4458 | 3154 | 3622 | 2047 |

TABLE 2.4.- QUANTIFICATION OF THE AIR SURVEILLANCE PROGRAM IN THE PERIOD 1966-1991.

| LOCATION | SAMPLES | ANALYSES | |
|---|---|---|---|
| | | Pu-239+Pu-240 | Am-241 |
| STATION P | 5630 | 960 | 245 |
| STATION 2 | 5884 | 924 | 163 |
| STATION 2-1 | 1542 | 383 | 55 |
| STATION 3-2 | 1060 | 106 | 0 |

* In compound samples

TABLE 2.5.- AVERAGE ANNUAL CONCENTRATIONS OF Pu-239+Pu-240 IN THE AIR DURING THE PERIOD 1966-1990.

| YEAR | CONCENTRATION AT THE STATION ($\mu$ Bq/m$^3$) | | | |
|---|---|---|---|---|
| | 2-1 | 2-2 | P | 3-2 |
| 1966 | 41,8 | 44,8 | 14,8 | 27,4 |
| 1967 | 15,2 | 441,8 | 4,1 | 13,0 |
| 1968 | 7,0 | 21,8 | 2,6 | 3,3 |
| 1969 | 161,0 | 142,0 | 2,6 | 14,1 |
| 1970 | | 5,9 | 2,2 | |
| 1971 | | 2,2 | < 1,8 | |
| 1972 | | 10,4 | < 1,8 | |
| 1973 | | 3,0 | 2,2 | |
| 1974 | | 8,1 | 4,1 | |
| 1975 | | 16,3 | < 1,8 | |
| 1976 | | 4,4 | < 1,8 | |
| 1977 | | 11,8 | 5,6 | |
| 1978 | | 16,7 | 2,2 | |
| 1979 | | 19,2 | 5,6 | |
| 1980 | | 32,9 | 28,1 | |
| 1981 | | 46,6 | 14,3 | |
| 1982 | | 60,4 | 18,6 | |
| 1983 | | 87,9 | 9,1 | |
| 1984 | 18,7 | 339 | 4,9 | |
| 1985 | 63,9 | 64,4 | 5,0 | |
| 1986 | 405 | 48,5 | 4,7 | |
| 1987 | 135,4 | 63,0 | 12,5 | |
| 1988 | 59,2 | 21,6 | 4,4 | |
| 1989 | 20,5 | 2,5 | 2,6 | |
| 1990 | 6,6 | 4,6 | 7,2 | |

TABLE 2.6.- QUANTIFICACION OF THE VEGETATION SURVEILLANCE PROGRAM IN THE PERIOD 1966-1991.

| CROP | SAMPLES | ANALYSES | |
|---|---|---|---|
| | | Pu-239+Pu-240 | Am-241 |
| Tomatoes | 454 | 769 | 428 |
| Barley | 771 | 1272 | 732 |
| Others | 723 | 1230 | 557 |

TABLE 2.7.- PLUTONIUM CONTAMINATION OF CROPS IN THE PERIOD 1966-1986.

| CROP | | SAMPLES | | Pu-239+Pu-240 Bq/kg | | |
|---|---|---|---|---|---|---|
| SPECIES | PART | TOTAL | % POSITIVE | AVERAGE | MAXIMUM | MINIMUM |
| Tomato | fruit | 236 | 9.74 | 0.03 | 1.05 | D.L |
| " | plant | 118 | 38.1 | 3.17 | 27.08 | D.L. |
| Barley | grain | 395 | 21.5 | 0.85 | 10.91 | D.L. |
| " | straw | 403 | 33.7 | 2.61 | 30.82 | D.L. |
| Maize | grain | 16 | 0 | 0 | 0 | D.L. |
| " | plant | 48 | 18.75 | 0.97 | 10.47 | D.L. |
| Bean | grain | 26 | 0 | 0 | 0 | D.L. |
| " | pod | 26 | 7.7 | 0.005 | 0.13 | D.L. |
| Alfalfa | | 78 | 42.3 | 0.60 | 4.33 | D.L. |
| Melon | fruit | 26 | 0 | 0 | 0 | D.L. |
| " | plant | 14 | 28.6 | 0.27 | 1.10 | D.L. |
| Fig | fruit | 24 | 8.3 | 0.03 | 10.61 | D.L. |
| Prickly Pear | fruit | 16 | 6.25 | 0.001 | 0.06 | D.L. |
| Pepper | fruit | 6 | 0 | 0 | 0 | D.L. |
| Orange | fruit | 6 | 0 | 0 | 0 | D.L. |
| Lemon | fruit | 6 | 0 | 0 | 0 | D.L. |
| Carob | fruit | 6 | 0 | 0 | 0 | D.L. |

DL. Detection Limit.

2.8.- QUANTIFICATION OF THE ANIMALS SURVEILLANCE PROGRAM IN THE PERIOD 1966-1991.

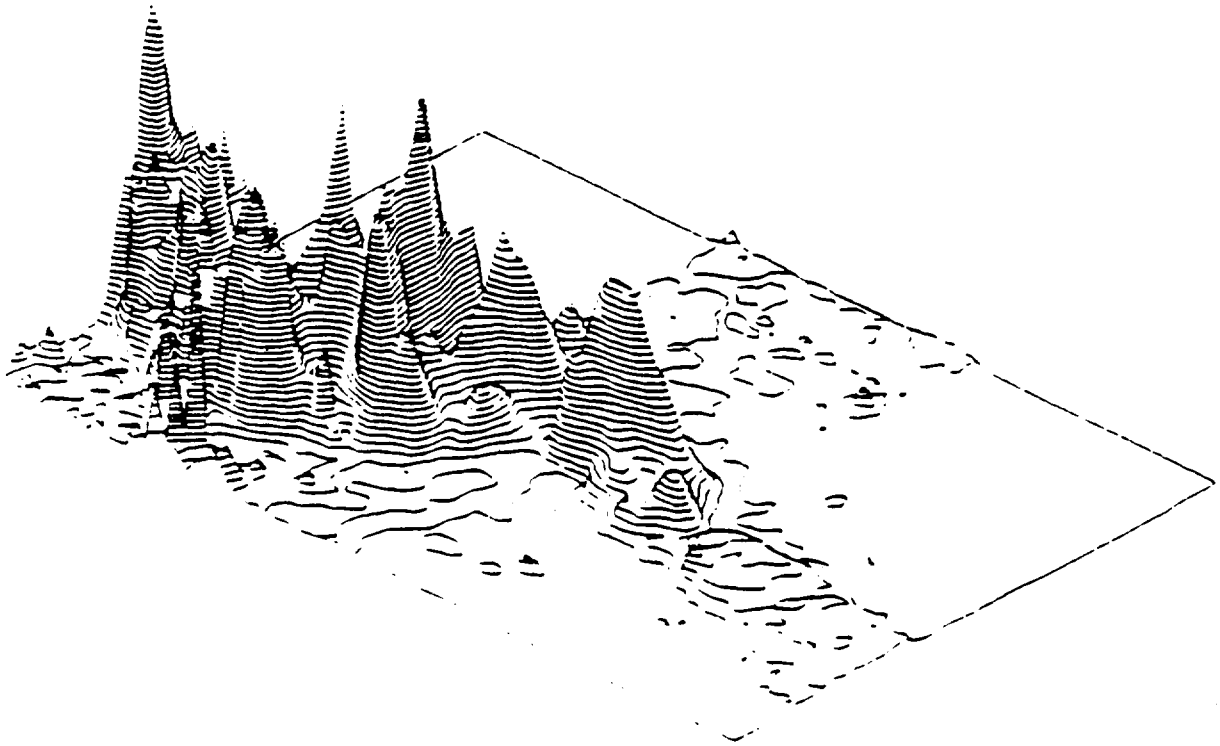
| ANIMAL | | SAMPLES | ANALYSES |
|---|---|---|---|
| SPECIE | TISSUE OR SECRETION | | Pu-239+Pu-240 |
| Snail | Shell | 15 | 15 |
| | Body | 15 | 15 |
| Goat | Milk | 27 | 27 |
| | Muscle | 2 | 2 |
| | Bone | 2 | 2 |
| Chicken | Meet | 10 | 10 |
| | Feathers | 10 | 10 |
| | Bone | 10 | 10 |
| | Gizzard | 10 | 10 |
| | Liver | 4 | 10 |
| | Kidneys | 4 | 10 |

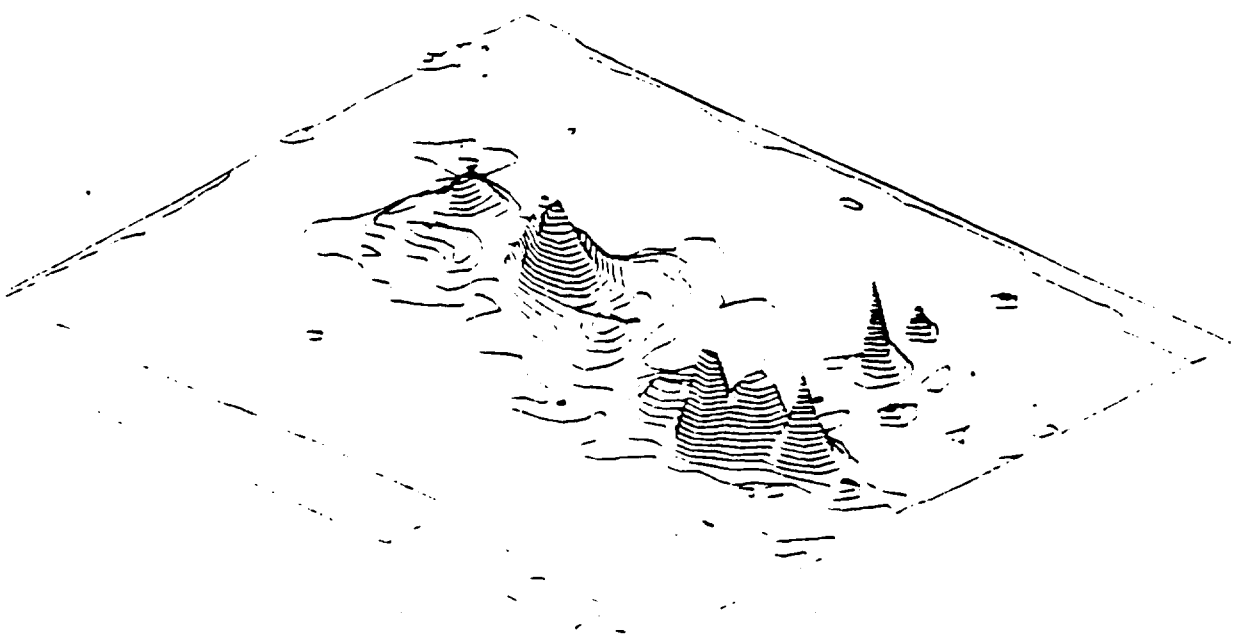
TABLE 2.9.- QUANTIFICATION OF THE PEOPLE SURVEILLANCE PROGRAM IN THE PERIO 1966-1989.

| EXAMINATIONS PER PERSON | EXAMINATED PEOPLE | | | EXAMINATION |
|---|---|---|---|---|
| | MALES | FEMALES | TOTAL | |
| 1 | 112 | 93 | 205 | 205 |
| 2 | 163 | 160 | 323 | 646 |
| 3 | 37 | 42 | 79 | 237 |
| 4 | 20 | 32 | 52 | 208 |
| 5 | 14 | 13 | 27 | 135 |
| 6 | 14 | 9 | 23 | 138 |
| 7 | 7 | 5 | 12 | 84 |
| 8 | 5 | 6 | 11 | 88 |
| 9 | 9 | 4 | 13 | 117 |
| 10 | 5 | 4 | 9 | 90 |
| 11 | 2 | 6 | 8 | 88 |
| 12 | 2 | 2 | 4 | 48 |
| 13 | 1 | 0 | 1 | 13 |
| 14 | 1 | 1 | 2 | 28 |
| TOTALS | 392 | 377 | 769 | 2125 |

TABLE 2.10.- PLUTONIUM EXCRETION IN URINE OF PEOPLE DURING THE PERIOD 1966-1991.

| ESCRETION IN URINE mBq/day | ANALYSES | PERCENTAGE | |
|---|---|---|---|
| | | PARTIAL | ACCUMULATE |
| ⟨ 0.37 | 1757 | 92.62 | 92.62 |
| 0.4-1.8 | 71 | 3.74 | 96.36 |
| 1.8-3.7 | 39 | 2.05 | 98:41 |
| 3.7-18.5 | 24 | 1.26 | 99.67 |
| 18.5-37.0 | 4 | 0.22 | 99.89 |
| 37.0-55.5 | 2 | 0.10 | 99.99 |





## 3. SOIL STUDIES

### 3.1 INTRODUCTION

The relative importance of inhalation and ingestion as pathways for human exposure depends on many enviromental parameters exclusive of the physicochemical associations of the radioelements in soils. Therefore in order to evaluate the real contamination risk to the people living in the area of Palomares is important to advance in the knowledge about the physicochemical association of plutonium and americium in the soil and the process of actinide-soil interactions. Furthermore theses parameters will influence the transport of this actinides , Resuspension and Vertical Migration and their behaviour on the human body.

Detritical calcareous-silicic soil samples from farmed and natural plots of Palomares have been taken to study the distribution of the transuranides and to follow their migration in depth (to 100 cm) on natural and agricultural environment.

The transuranide concentrations have been determined in relation to depth, particle size distribution and mineralogical composition of the soil. Secuential leaching experiments and authoradiographical techniques have been used to estimate the chemical distribution and behaviour of these radionuclides.

### 3.2. CHEMICAL AND MINERALOGICAL COMPOSITION OF THE SOIL

A phisico-chemical characterization of the soil has been made in order to define the physico-chemical environment that determine the stability of the mineral components and the state and therefore stability of the radionuclides that this soil content.

The chemical composition has been studied on the total sample and the

differents size particles fractions. Cation and anion macroconstituents were determined.

The techniques used were :

a) Induced coupled plasma-optical emission spectroscopy
b) Ionic cromatography and specific electrode
c) Turbidimetry
e) Flucrimetry
f) Elemental analyser to determine carbon and sulphur.

According to the petrographic composition, the samples are classified as lithograywacke-phylloarenite.

The chemical contents, of the total sample and the granulometric fractions, are shown in the table 3.1

table 3.1 Chemical composition of the total sample and of the granulometric fractions

| Concent. % | Total sample | >1000 µm | 1000–250 µm | 250–125 µm | 125–63 µm | 63–40 µm | 40–20 µm | 20–10 µm | 10–5 µm | <5 µm |
|---|---|---|---|---|---|---|---|---|---|---|
| $SiO_2$ | 44.0 | 49.7 | 49.8 | 60.6 | 52.5 | 44.4 | 44.1 | 42.1 | 39.0 | 34.3 |
| $Al_2O_3$ | 12.5 | 5.0 | 3.1 | 9.8 | 11.6 | 10.2 | 13.9 | 15.6 | 15.2 | 13.4 |
| CaO | 14.3 | 11.4 | 15.0 | 10.2 | 9.4 | 13.5 | 13.6 | 14.7 | 14.7 | 16.4 |
| $Fe_2O_3$ | 3.76 | 9.43 | 4.87 | 2.35 | 2.71 | 4.16 | 3.91 | 4.05 | 4.11 | 4.75 |
| FeO | .66 | .51 | .46 | .49 | .53 | .44 | .62 | .76 | .71 | .58 |
| MgO | 2.1 | 1.1 | .91 | 1.3 | 1.5 | 1.8 | 2.4 | 2.7 | 2.6 | 2.5 |
| MnO | .047 | .047 | .076 | .046 | .039 | .044 | .046 | .049 | .053 | .055 |
| $K_2O$ | 2.6 | .60 | .72 | 1.5 | 1.9 | 1.7 | 2.9 | 3.5 | 3.9 | 3.6 |
| $Na_2O$ | .96 | .47 | .43 | .58 | .52 | .62 | .84 | .96 | .37 | .56 |
| $TiO_2$ | .60 | .25 | .25 | .50 | .64 | .96 | .68 | .60 | .52 | .46 |
| $P_2O_5$ | .15 | .11 | .11 | .09 | .12 | .19 | .17 | .16 | .16 | .17 |
| $CO_3^{=}$ | 15.39 | 8.49 | 8.89 | 7.59 | 9.39 | 14.39 | 16.44 | 18.34 | 20.58 | 23.58 |
| C. org | .22 | .10 | .22 | .12 | .12 | .12 | .21 | .33 | .28 | .28 |
| $SO_4^{=}$ | 1.1 | 6.1 | 7.2 | 1.2 | .18 | .51 | .20 | .15 | .14 | .17 |
| $S^{=}$ | .26 | .36 | 2.10 | .15 | .00 | .09 | .00 | .03 | .03 | .01 |
| $Cl^-$ | .20 | .02 | .02 | .02 | .02 | .04 | .06 | .08 | .09 | .11 |
| $NO_3^-$ | 48 | <1 | <1 | 54 | 84 | 78 | 36 | 30 | 50 | 45 |
| U | 3.7 | 2.2 | 3.7 | 5.5 | 4.2 | 6.0 | 4.2 | 4.2 | 4.2 | 5.5 |
| Ba | 420 | 130 | 210 | 250 | 310 | 270 | 475 | 580 | 550 | 450 |
| Co | 15 | 13 | 12 | 11 | 10 | 15 | 17 | 18 | 17 | 18 |
| Cr | 61 | 190 | 40 | 37 | 39 | 63 | [illegible] | 36 | 34 | 30 |
| Cu | 23 | 22 | 50 | 12 | 10 | 15 | 23 | 27 | 30 | 27 |
| La | 28 | 11 | 16 | 14 | 22 | 34 | 49 | 48 | 46 | 46 |
| Li | 49 | 18 | 22 | 31 | 34 | 32 | 55 | 57 | 59 | 56 |
| Ni | 28 | 135 | 21 | 21 | 20 | 28 | 37 | 40 | 39 | 46 |
| Pb | 49 | 46 | 32 | 30 | <25 | 30 | 43 | 57 | 63 | 33 |
| Sr | 575 | 250 | 315 | 565 | 540 | 630 | 455 | 475 | 545 | 620 |
| V | 32 | 42 | 42 | 49 | 60 | 73 | 93 | 105 | 105 | 105 |
| Zn | 58 | 35 | 60 | 62 | 42 | 98 | 21 | 150 | 155 | 210 |
| Zr | 199 | 97 | 71 | 101 | 147 | 545 | 259 | 174 | 123 | 35 |

Based on its chemical mineralogical composition, the soil has a siliceous-carbonatic nature. Therefore , $SiO_2$, $Al_2O_3$, CaO and $CO_3^=$ are its main components.

The $Fe^{2+}/Fe^{3+}$ ratio is 0.18 for the total sample, reaching its lowest value ,0.05, in the fraction with sizes greater than 1000μm. This relation indicates an oxidizing enviromment wich is in good agreement with the detected mineralogical alteration-transformations, biotites and chlorites degradation and surface alteration of iron and titanium minerals.

The content of $SO_4^=$ is the most significant among the most soluble components. It is found as gypsum in the granulometric fractions with sizes greater than 250μm.

The Techniques used to carried out the mineralogical analyses were :

- Optical microscopy
- X-Ray Diffraction

The mineralogical composition of the main components of the soil is shown in table 3.2

table 3.2 Mineralogical composition of the total sample and of the granulometric fractions

| Concent. % | Total sample | >1000 μm | 1000–250 μm | 250–125 μm | 125–63 μm | 63–40 μm | 40–20 μm | 20–10 μm | 10–5 μm | <5 μm |
|---|---|---|---|---|---|---|---|---|---|---|
| Quartz | 38.61 | 51.38 | 54.77 | 59.75 | 47.39 | 55.78 | 28.97 | 17.10 | 12.45 | 10.28 |
| Mica | 4.06 | 0.43 | 0.36 | 0.71 | 4.89 | 1.20 | 5.19 | 5.89 | 7.52 | 7.78 |
| Chlorite | 5.52 | 1.72 | 3.21 | 4.99 | 4.89 | 3.72 | 6.55 | 7.86 | 9.14 | 6.67 |
| Plagioclase | 13.46 | 13.22 | 5.57 | 15.69 | 12.29 | 6.97 | 12.42 | 16.94 | 15.66 | 10.74 |
| Orthoclase | 0.31 | tr | tr | 1.90 | 0.84 | 1.39 | 1.35 | 2.37 | 2.03 | 2.31 |
| Calcite | 30.09 | 10.40 | 14.13 | 7.92 | 27.42 | 20.92 | 30.31 | 38.71 | 46.49 | 57.69 |
| Dolomite | 6.97 | 3.61 | 6.42 | 6.18 | 1.96 | 7.77 | 14.15 | 10.72 | 6.60 | 4.07 |
| Gypsum | 0.49 | 13.74 | 15.55 | 2.85 | 0.33 | 2.26 | 0.53 | 0.41 | tr | 0.46 |

This composition is relatively simple in relation with the main components but is rather complex concerning to the number of neoformed and inherited minerals. The proportion of the latter is not significant.

The dominating minerals are quartz, representing 38,6% of the sample and carbonates with 37%, being Calcite, the main component of the rest of microforaminifera fossils in the soils the dominant. The dolomite content is 7%.

The general tendency of the variation of the mineralogical composition of the granulometric fractions consits of a decrease in quartz and feldespars and an increase in carbonates and phyllosilicates as the grain size decreases. Gypsum occurs generally in sizes greater than 250μm

Some physico-chemical parameters as pH , Eh and conductivity has been measured on the water soil solution of soil samples taken from differents plots . Eh varies from 312 to 344 mV and pH from 7.6 to 8.43. This values of the samples indicate an oxidizing and slightly alkaline enviroment.

According to the Eh-pH diagram it would seem that we are in the theoretical region of stability of $PuO^2$, althought the equilibrium concentration with $PuO^{2+}$ is higher than the teoretical value. This seems to indicate the presence of other chemical species.

The chemical composition of this soil solution has been also determined . The table 3.3 show these results.

Table 3.3 Ionic content on water soil solution

| Ions / mg/l | A.2-0 | A.2-1 | A.2-2 | A.3-1 | A.3-2 | A.5-1 | A.5-2 | A.5-3B |
|---|---|---|---|---|---|---|---|---|
| $CO_3H^-$ | 170,15 | 383,56 | 323,58 | 111,82 | 115,20 | 239,38 | 165,15 | 83,79 |
| $CO_3^{2-}$ | - | - | - | - | - | - | - | - |
| $Cl^-$ | 18,94 | 59,33 | 444,22 | 729,14 | 421,51 | 2538,38 | 2038,41 | 1448,76 |
| $NO_3^-$ | 4,97 | 3,31 | 4,44 | 287,70 | 79,01 | 1832,40 | 128,30 | 224,52 |
| $NO_2^-$ | 1,54 | 1,52 | 0,46 | 24,75 | 19,50 | 295,52 | 0,26 | 7,43 |
| $PO_4^{3-}$ | < 2,00 | 4,17 | 4,68 | < 2,00 | < 2,00 | < 2,00 | < 2,00 | < 2,00 |
| $SO_4^{2-}$ | 92,36 | 166,76 | 125,08 | 1739,58 | 549,48 | 2721,32 | 269,30 | 2506,51 |
| $Na^+$ | 56,83 | 44,30 | 202,25 | 314,49 | 211,11 | 935,70 | 675,14 | 314,95 |
| $K^+$ | 73,41 | 77,75 | 73,65 | 99,01 | 53,68 | 85,77 | 109,06 | 106,36 |
| $Ca^{2+}$ | 36,71 | 57,32 | 37,41 | 297,02 | 470,47 | 717,37 | 519,34 | 516,99 |
| $Mg^{2+}$ | 8,54 | 17,72 | 39,75 | 206,75 | 111,59 | 409,37 | 231,11 | 115,88 |
| $Fe^{2+}$ | <0,10 | <0,10 | <0,10 | <0,10 | <0,10 | <0,10 | <0,10 | <0,10 |
| $Al^{3+}$ | <0,50 | <0,50 | <0,50 | <0,50 | <0,50 | <0,50 | <0,50 | <0,50 |

## 3.3 ACTIVITY CONCENTRATION DISTRIBUTION IN FUNCTION OF PARTICLE SIZE

An evaluation of the association of plutonium in the differents size of soils particles has been done . This is important in order to know the potential hazard due to resuspension and inhalation of contaminated particles and the behaviour of the radionuclides associated to this soil.

The samples were subjected to maceration and ultrasonic dispersion. From each sample, eight size fractions were obtained within the following size intervals :

1000-250-125-63-40-20-10-5-0-μm

Fractional segregation of soil particles greater than 63μm, after leaching with water over a mesh of a prescribed size, was carried out by screening according to NLT 104/72 Standars (SMTL72).

Soil particles smaller than 63 μm were fractionated by the "British Rema" air classifier. Soil density was considered to be 2.7 g x $cm^{-3}$.

The soil size distribution is polimodal and comprises from gravel to clays. However, the silt-size components ( 63 > S > 5μm) predominate with 55% (Iranzo et al. 1990).

Plutonium analyses has been performed in the fractions isolated. Table 3.4 shows the distribution of the concentration of Pu-239+Pu-240 among the different granulometric fractions.

**Table 3.4. Plutonium activity in the particles size fractions of soil**

| Size class μm | Abundance % | $^{239}Pu - ^{240}Pu$ | |
|---|---|---|---|
| | | Bq · g$^{-1}$ | Abundance % |
| $S < 5$ | 8.781 | 504 ± 14 | 2.32 |
| $5 < S < 10$ | 11.400 | 1333 ± 79 | 7.96 |
| $10 < S < 20$ | 8.772 | 1056 ± 67 | 4.85 |
| $20 < S < 40$ | 23.730 | 3370 ± 210 | 41.87 |
| $40 < S < 63$ | 10.765 | 1504 ± 82 | 8.48 |
| $63 < S < 125$ | 13.788 | 2183 ± 99 | 15.76 |
| $125 < S < 250$ | 4.600 | 6389 ± 315 | 15.39 |
| $250 < S < 1000$ | 2.374 | 904 ± 42 | 1.36 |
| $1000 < S$ | 15.192 | 254 ± 9 | 2.02 |

The average $^{239+240}$ Pu concentration in the total sample is 1944 Bq.g$^{-1}$. The concentration values range from 254 to 6389 Bq.g$^{-1}$. The bimodal distribution curve shows modal values of 3370 Bq.g$^{-1}$ between 40 and 20 μm, and 6389 Bq.g$^{-1}$ from 250 to 125 μm, wich represent, respectively, 41.9 and 15.4 % of the total $^{239+240}$ Pu content

The concentration and content of Pu in the granulometric fraction, ranging from 250 to 125 μm should be emphasized, because it bears 15,4% of the total Pu, and represents only 4,6% of the sample. Due to the high concentration of Pu in this fraction (6389 Bq/gr) it has been considered of great interest to study its distribution and its chemical-mineralogical association with the other components (Iranzo et al. 1990).

This fraction has been divided in 16 fractions using densimetric and magnetic techniques. Chemical and Mineralogical composition and Plutonium distribution has been determined (E.Iranzo et all 1991).

In general Pu concentration increases with the density and with the magnetic class. This increase with the magnetic susceptibility is shown in the two fractions with density lower than 3.3 g.cm$^{-1}$, but in the fraction with density higher than 3.3 g.cm$^{-1}$ the maximum concentration is reached

with the lowest magnetic susceptibility. Just this fraction, that consits of titanium minerals (ilmenite, rutile, sphene), iron minerals (maghemite, maghemite and hematite), pyroxenes of the enstatite-hypersthene series as well as angite and garnets of the grossularite-andradite and spessartite-pyrope series, contain the highest Pu concentration (502,5 KBq/g )

## 3.4 GEOCHEMICAL ASSOCIATION OF PU AND AM

The Knowledge of the geochemical association of plutonium and americium in the Palomares soil came from three differents areas of studies:

a) Interpretation of the data obtained from the chemical and mineralogical studies.

b) Sequential leaching studies.

c) Autorradiographie studies.

A) Data interpretation. As it has been indicated from the chemical and mineralogical composition studies the Palomares soil basically corresponds to the C horizon formed by disgregation and metheoric alteration of the underlying sediments, and has a siliceous - carbonatic nature (Iranzo et al. 1990).

It is noteworthy that the plutonium is associated to the $Fe^2O^3$ in the density fraction < 2.8 $g.cm^{-3}$, practically lacking in cristalline iron minerals. This is interpreted in the sense that the neoformation iron hydroxides have co-precipitated part of the mobilizable plutonium.

The association of the plutonium with the carbonates is evident in the density fraction >3.3 $g.cm^{-3}$. In the density fraction > 2.8 $g.cm^{-3}$, it is associated to the neoformation dolomite.

B) Sequential leaching studies. In order to gain further knoledge of the geochemical associations that Pu and Am show in the soil, a sequential leaching method has been applied.

The methodology emploied is the secuential leaching method firstly developed by Mc Laren and Crawford (1973) and subsequently modified to applicate it in geochemical studies on transuranics in contaminated soils (Livens et al. 1986). We have applied this methodology including some modifications brougth in by our work group (Iranzo et all 1991 and Aragón et al. 1992) and its shown in table 3.5.

These extraction techniques were designed to identify the percenteages of plutonium and americium in the following forms :

Readily avaible, exchangeable and bound to specific inorganic adsorption sites, associated with organic matter as chelated complexes, adsorbed onto or co-precipitated with sesquioxides (aluminium, manganese and iron) and finally a residual fraction. We have included a first step with deionized water and also we divide the residual fraction between the fraction directly extracted by concentrated nitric acid and the other extracted by 12 M $HNO_3$/1M HF after ashing.

In addition, we have been doing some test with the aim to determinate the necesary time to reach chemical equilibrium soil-extractant. In relaction with this we have developed the equilibrium curve activity - time, making a theoretical approximation. A noteworthy pint is the reduction in the activity extracted with the passing of time in the case of distilled water and $CaCl_2$ (fig.3.1). This fact may be due to the formation of colloids of a size greater than 0.45µm which would be retained while the solutions are being filtered.

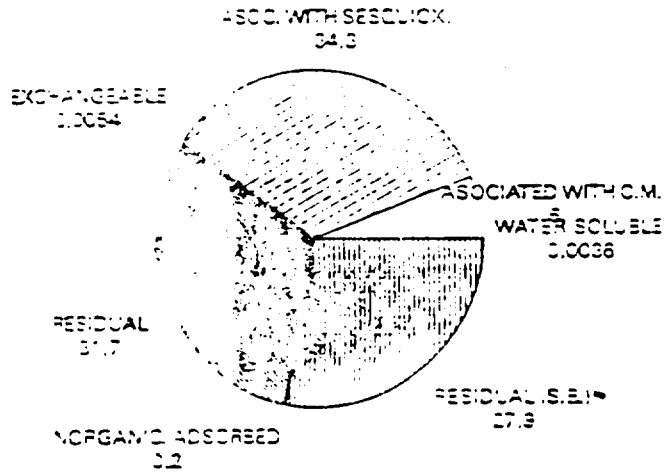
One similar fact ocurred during alkaline leaching experiences, in which we can see a reduction in the activity extracted with the passing of time in the case of NaOH $10^{-2}$N (P. Rivas, 1991).

Table 3.5 Secuential Leaching methodology applied in this work

| FRACTION | REAGENT | VOLUME (ml/gr) |
|---|---|---|
| WATER SOLUBLE | DISTILLED WATER | 20 |
| EXCHANGEABLE | 0.05M $CaCl_2$ | 20 |
| SPECIFICALLY ADSORBED | 0.5M ACETIC ACID | 20 |
| ORGANICALLY BOUND | 0.1M $Na_4P_2O_7$ | 100 |
| OXIDE BOUND | 0.175M $(COONH_4)_2$ | |
| | 0.1M $(CO_2H)_2$ | 75 |
| RESIDUAL | 7.5M $HNO_3$ | 70 |
| RESIDUAL (STRONGLY BOUNDED) | HF - $HNO_3$ | 70 |

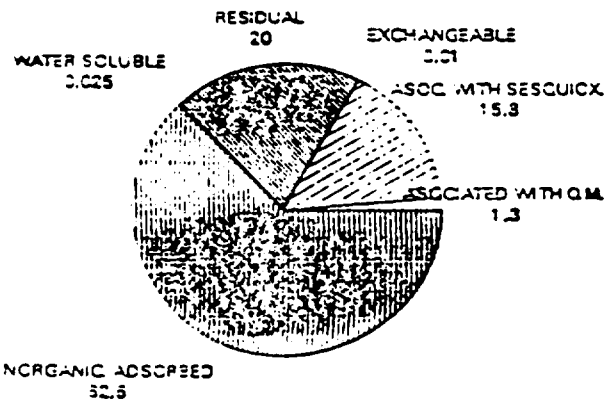
The results obtained on $Pu^{239}$ + $Pu^{240}$, $Pu^{238}$ and $Am^{241}$ in the diferent soil fractions are shown in the following figures :

*PERCENTAGE OF Pu-239 + Pu-240 CONTENT IN THE OBTAINED FRACTIONS*




*MEAN VALUES*

*PERCENTAGE OF Am-241 CONTENT IN THE OBTAINED FRACTIONS*




*MEAN VALUES*

From this data ( Iranzo et all 1991, Aragon et all 1992), it can be deduced that :

. Approximately 93 per cent of the activity concentration of Pu-239 + Pu-240 is found to be associated with sesquioxides and distributed between the two residual fractions, and probably incorporated in the crystalline structure of resistant minerals ( chlorite,biotite), complex oxides of Ti, Fe, Mn and coprecipitated or in diadoxic substitution with carbonates (dolomite, ankerite) and phosphates.

. The most mobile compounds of Pu represent only a small fraction, 0.21%, of the total Pu.

. Aproximately 6% is fixed on organic matter

. The Pu-238 distribution does not vary substantially from that of the Pu-239, except in the soluble and exchangeable fractions, where the percentage of Pu-238 is around three times the Pu-239 percentage.

. Approximately 63 per cent of the Am extracted is found in the greatest mobility phase, made up of the soluble, exchangeable and inorganically-adsorved fractions.

. The greatest mobility phase of the americium is much higher than the greatest mobility phase of the plutonium.

. The formation of colloids may occur during extraction.

C) **Autorradiographie studies.** These studies has been done in the samples that present the highest alpha activity. These autorradioghaphies were obtained on plastic, type CR-39 (oxydi-2,1-ethanedyl di-2-propenyl diester of carbonic acid), placed over thin polished sections of the granulometric fractions during 32 days.

Autorradiographies show different concentration levels in different suported minerals (E. Iranzo et all 1986).

## 3.5 VERTICAL MIGRATION STUDIES

The vertical distribution of Pu in soil has been studied in soils profiles corresponding to cultivated and non-cultivated soil.

The results (tables 3.6 and 3.7) shows that in non-cultivated soils, more than 99% of Pu is found in the first 5 cm of soil; however a certain level of contamination has been measured in all the profiles and it decreases continuously down to 45 cm.

The non cultivated soil zone (table 3.6), in wich the sampling were done, is located uphill with the streams converging toward the cultivated-soil-zone. This soil profile, not disturbed by ploughing activities, has been taken as reference to make an stimation of the retardation factor.

Table 3.6 plutonium activity in soil profiles of non cultivated soils

| Profile | Depth cm | $^{239}Pu - ^{240}Pu$ Bq × g$^{-1}$ | Abundance % |
|---|---|---|---|
| 31 | 0– 5 | 382 ±57 | 99.6029 |
| 31 | 5–15 | 1.43 ± 0.30 | 0.3729 |
| 31 | 15–25 | 0.050 ± 0.008 | 0.0130 |
| 31 | 25–35 | 0.032 ± 0.005 | 0.0083 |
| 31 | 35–45 | 0.011 ± 0.002 | 0.0029 |
| 33 | 0– 5 | 43.9 ± 6.6 | 99.5804 |
| 33 | 5–15 | 0.076 ± 0.014 | 0.1724 |
| 33 | 15–25 | 0.077 ± 0.013 | 0.1747 |
| 33 | 25–35 | 0.030 ± 0.006 | 0.0681 |
| 33 | 35–45 | 0.002 ± 0.001 | 0.0045 |
| 35 | 0– 5 | 461 ±69 | 99.4995 |
| 35 | 5–15 | 2.31 ± 0.35 | 0.4986 |
| 35 | 15–25 | 0.004 ± 0.001 | 0.0009 |
| 35 | 25–35 | 0.003 ± 0.001 | 0.0006 |
| 35 | 35–45 | 0.002 ± 0.001 | 0.0004 |
| 36 | 0– 5 | 471 ±71 | 99.9400 |
| 36 | 5–15 | 0.042 ± 0.007 | 0.0089 |
| 36 | 15–25 | 0.024 ± 0.0004 | 0.0051 |
| 36 | 25–35 | 0.010 ± 0.002 | 0.0021 |
| 36 | 35–45 | 0.007 ± 0.002 | 0.0015 |
| 37 | 0– 5 | 263 ±39 | 99.3908 |
| 37 | 5–15 | 1.29 ± 0.19 | 0.4875 |
| 37 | 15–25 | 0.089 ± 0.013 | 0.0336 |
| 37 | 25–35 | 0.023 ± 0.004 | 0.0087 |
| 37 | 35–45 | 0.010 ± 0.002 | 0.0038 |

In the downstream cultivated soils, a homogenization of the concentration is observed , due to the crops, as well as a decrease of the concentration with depth. However, the lowest concentration levels are higher tham the lowest in the uphill profile.

On the other hand, it must be emphasized the existence in the cultivated soil profiles of an accumulation level at a depth of between 70 and 90 cm deep. It can be determined in one of the profile by a minimum in soil porosity and permeabililty and in the other profile by an increase in the organic matter content, wich has probably acted as a accumulation reducing barrier.

Table 3.7 plutonium activity in soil profiles of farming soils

| Depth cm | $^{239}Pu - ^{240}Pu$ activity Bq × g$^{-1}$ | | $^{239}Pu - ^{240}Pu$ abundance % | |
|---|---|---|---|---|
| | Profile 2-0 | Profile 2-2 | Profile 2-0 | Profile 2-2 |
| 0 – 5 | 4700 ± 705 | 18.4 ± 2.8 | 97.63 | 17.45 |
| 5 – 10 | 40.1 ± 6.0 | 18.0 ± 2.8 | 0.83 | 17.07 |
| 10 – 20 | 27.0 ± 4.5 | 17.5 ± 2.7 | 0.56 | 16.59 |
| 20 – 30 | 10.4 ± 1.6 | 2.6 ± 0.40 | 0.22 | 2.47 |
| 30 – 40 | 4.25 ± 0.64 | 0.16 ± 0.03 | 0.09 | 0.15 |
| 40 – 50 | 0.0778 ± 0.0170 | 0.024 ± 0.005 | 0.002 | 0.02 |
| 50 – 60 | 6.6 ± 1.0 | 4.70 ± 0.72 | 0.14 | 4.46 |
| 60 – 70 | 0.3434 ± 0.0549 | 0.02 ± 0.004 | 0.007 | 0.02 |
| 70 – 80 | 23.0 ± 3.5 | 0.05 ± 0.007 | 0.48 | 0.05 |
| 80 – 90 | 2.14 ± 0.32 | 35.30 ± 5.44 | 0.04 | 33.96 |
| 90 – 100 | 0.0298 ± 0.0081 | 0.040 ± 0.006 | 0.0006 | 0.04 |
| 100 – 110 | | 0.09 ± 0.016 | | 0.09 |
| 110 – 120 | | 7.94 ± 1.210 | | 7.53 |

### 3.5.1. Conclusions

. Taking into account an estimation of the migration parameters of plutonium in these soils: time, 23 years; penetration depth, 40cm; soil permeability, $2.02*10^{-7}$cm/s, the retardation factor is assume to be 2.93, and the distribution coefficient aproximately 0.3 ml/gr.(Irazo et all 1990).

. The vertical migration speed of plutonium in the soil of Palomares is in the order of $10^{-8}$ cm/s. (E.Iranzo et all 1990).

. Variations on the hydraulic parameters and Oxi-reduction conditions can produce some accumulation levels.

## 3.6 FUTURE STUDIES

* To advance in the vertical migration studies. A new technique in order to obtain the profile will be used. The results obtained will be compared.

* Study of the accumulations levels and the process that produce them.

* Edaphologic studies

* To advance the studies about geochemical association of transuranides elements. Studies about association of Americium to soil particles. Comparison of its behaviour with plutonium behaviour.

* Studies about colloids forms.

* Hot particles studies.

FIG.3.1 a

EXTRACTION WITH DISTILLED WATER

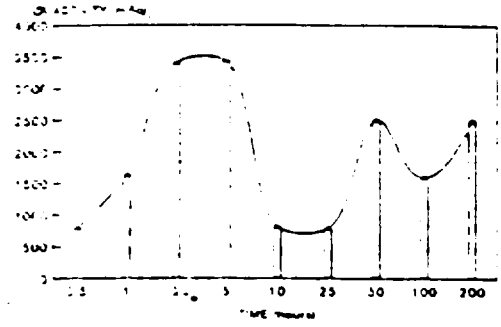


FIG.3.1 b

EXTRACTION WITH $CaCl_2$ 0.05M

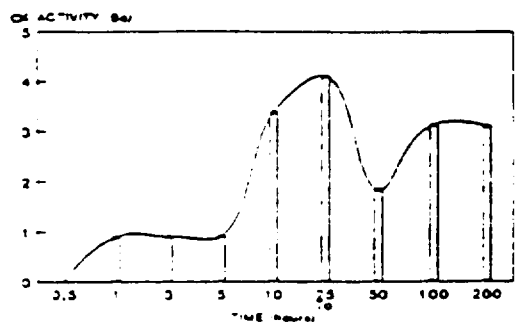


FIG.3.1 c

EXTRACTION WITH ACETIC ACID 0.5M

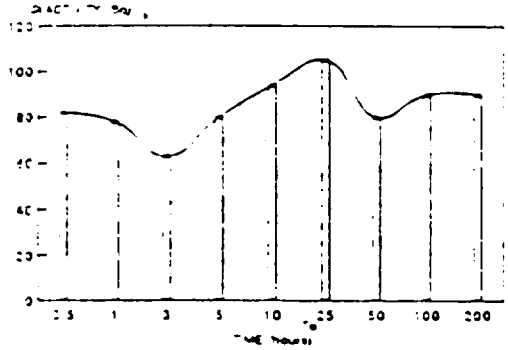


FIG.3.1 d

EXTRACTION WITH SODIC PIROFOSFATE 0.1M

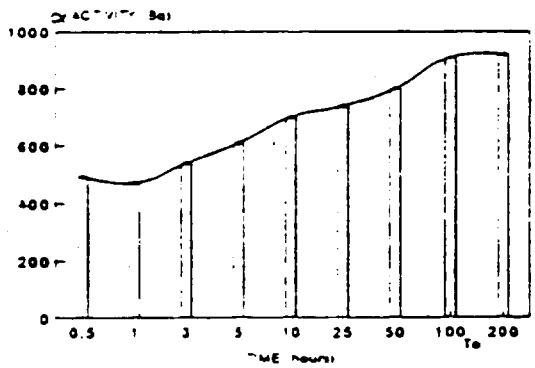


FIG.3.1 e

EXTRACTION WITH AMONIUN OXALATE 0.175M

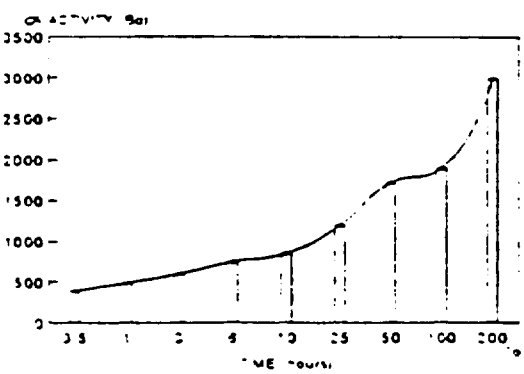
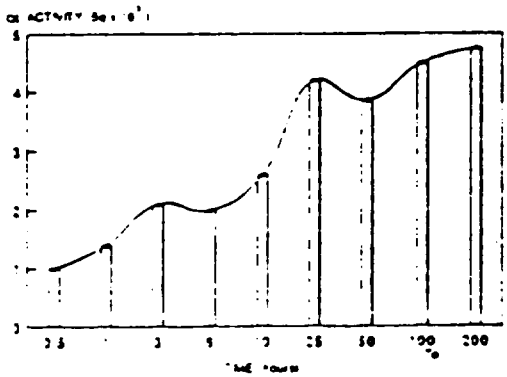


FIG.3.1 f

EXTRACTION WITH NITRIC ACID 7.8M



## 4. RESUSPENSION STUDIES

### 4.1 INTRODUCTION

Resuspension has been recognized as a long term potential mode of human exposure to contaminants in the soil. Radioactive material deposited onto the ground can be resuspended into the air by wind or by other disturbances. Also resuspension and subsequent re-deposition in agricultural areas may lead to contamination of crops and foods derived from grazing animals.

The inhalation pathway is the more important potential contamination pathway in the area of Palomares. It can be explain in one hand because of the climatology of the area, a typical agricultural Mediteranean area with scarce yearly precipitation that favours this pathway. In the other hand because the main source of contamination is the soil.

A better undestanding of the Resuspension processes will help in an important way to make predictions. Some resuspension parameters used to get this objective are defined in the following text.

### 4.2 RESUSPENSION PARAMETERS. DATA REVIEW.

Considering wind-driven resuspension, a review of the data obtained along the experimental radiological surveillance programme carried out in the area of Palomares since the time of the accident has allowed us to measure the following resuspension parameters :

. RESUSPENSION FACTOR, defined as the ratio of the concentration in the air at a reference heihgt to the quantity of the contaminant per unit area on the surface of the ground.

Resuspension factor has been calculated based on yearly Pu air concentration averages obtained along the years and concentration of Pu in surface soil ( E.Iranzo and Salvador 1970, E.Iranzo et al 1987 and E.Iranzo et al 1988). The data obtained (C.E. Iranzo et al, to be published) indicate that this factor decrease from an initial average value in the order of $10^{-7}$ to values in the order of $10^{-9}$ some month later and in the order of $10^{-10}$ years later. The interannual variability is about 40 % and the spacial variability even higher in a distance scale of hundred to thousand meters. Further studies are underway to confirme and refine this experimental evidences.

The data obtained in this mediterranean southern conditions are in good agreedment with the model of time dependent resuspension factor developed by Garland (Garland et al. 1990) based in northern European conditions.

. DUST LOADING, defined as ratio between activity concentration in the air and volumetric concentration in soil.

$$Se\ (Kg/m^3) = \frac{Ca\ (Bq.m^{-3})}{Cs\ (Bq.Kg^{-1})}$$

It has the advantage that it is not necessary to decide about the depth to consider.

Dust loading has been calculated based in weekly air concentration measurements taken during two consecutive years in two different stations located in Palomares area, and average soil concentration in the study plots. This data show an average around 100 $\mu g/m^3$ with a standard desviation higher than the average.( C.E.Iranzo et al, to be published ).

. RESUSPENSION RATE, defined as the fraction of the contaminant present on the ground that is resuspended per unit time .

$$TR(S^{-1}) = \frac{\text{Resuspension flow } (Bq.m^{-2}.s^{-1})}{\text{S sup } (Bq.m^{-2})}$$

Once it has been obtained it can be used to describe concentrations in some determined point of a inhomogeneus contaminated area using deposit and difusion ecuations and doing integrals over the area. It can be obtained from measurements executed in wind tunnels and laboratories studies, but it can not be easily deduced from field studies.

Based in this resuspension rate approach, a compartimental model that deals with heterogeneously contaminated soils, is being aplied. (García-Olivares et al 1992).

. MASS LOADING ONTO PLANT SURFACES.

The application of this approach to barley samples collected in 1983, has show the following values :

Barley grain ..... 53.3 mgr x $gr^{-1}$
Barley straw ..... 112.2 mgr x $gr^{-1}$
Espicule ..... 121.2 mgr x $gr^{-1}$

The highest value correspond to the part of the plant wich higher posibilities of surface retention of particles.

In relation with **Man-made Resuspension**, dust loading values up to $12mg/m^3$ has been calculated from a distance about 100m from the place where a very big pool for irrigation purposes was constructed using machines.

This data suggest that significantly higher values were probably to be found at the workplace. ( C.E. Iranzo, A. Espinosa, personal comunication).

## 4.3 REDISTRIBUTION of CONTAMINATION BY RESUSPENSION

To elucidate the role of the resuspended particles as vectors for the spread of deposited activity and try to measure the rate of spread of contamination into a previous clean area is an important aim in the resuspension studies. This fact can decrease in an important way the efectivity of some countermeasures some time after their application and it is an important pathway of secondary terrytory contaminations.

With this aim surface soil samples have been taken at 29 points located on the outside perimeter of what was identified as the "zero line" of initial contamination, at an approximate distance of 500m and 1000m from the latter. Figure 2.1 shows the original levels of contamination where the curve enclosing the area is the so called initial zero line.

Concentration values of Pu-239+Pu-240 activity higher than the minimum detectable concentration with our method of analysis (1.9 mBq x $g^{-1}$) were found in several samples corresponding to points situated 500m and 1000 m outside the zero line.

These data show a movement of particles beyond the contamination limit line after the accident in 1966. Resuspension is obviously one of the processes contributing to the redistribution of the initial contamination.

Furthermore, this redistribution of the contamination is confirmed by the appearance of activity concentration values higher than the Minimun activity concentration detected by our method of analysis in some samples of vegetation cultivated in a plot initially taken as a reference plot.

## 4.4 CURRENT STUDIES

Predicting the resuspension phenomenon is still a difficult task due

to the great number of factors involved, among which we may mention :

- Wind speed.
- Nature of the surface.
- Time elapsed since the deposit.
- Surface humidity.
- Size of the contaminating particles
- Soil chemistry and texture.
- Chemical properties of the contaminating agent.
- Deposit processes.
- Farming methods.
- Intensity and Frecuency of rainfall.
- Mechanical disturbance.

Many of these factors are characterized in the Palomares area, wich means an extremely value aid when interpreting the experimental results obtained.

In order to gain greater knowledge of the resuspension processes and to be able to determine their range, a test has been designed for the purpose of providing data on the resuspension and depositing of particles under natural conditions. The aim of this is to obtain a tracer concentration gradient in terms of particle height and size under well-defined meteorological conditions. Attention was initially focused on particles < 10 μm. In order to achieve this goal, high-volume samplers equipped with multistate cascade impactors have been installed at heights of 1.7 and 3 metres.

This experiment, as was said previously, is performed under defined meteorological conditions . An automatic station is provided with capacity for 16 analog input channels and data collection by means of storage inputs. The station is equipped with three triaxial anemometers, two temperature gauges and a relative-humidity and ambient-temperature gauge.

There are three measurement levels. A triaxial anemometer has been installed in each of them thereby allowing the vertical speed profile to be

obtained, as well as its three components, which makes it possible to determine the corresponding wind directions. The temperature gauges have been installed at two levels, and the relative humidity and ambient temperature gauge at the third. This situation makes it possible to obtain the vertical temperature profile with three measurements points and the corresponding gradients, which give us information about stability conditions and, at one level,relative humidity. It provides data on a continuous basis.

This study was preceded by a good characterization of local meteorology including a far-reaching study of breezes since it is a well known fact that southern European coastal regions are influenced by breeze cycles. (Aceña B, 1990 and 1991)

At the same time some work has been done in relation with models. A model of resuspension and air transport of particles has been created to be applied in this scenario. It is a compartimental model taking into account the resuspension, deposition and advective dispersion of soil particles.

In this model the whole region of interest is represented as a set of elemental cells. The balance is solved for steady state wind conditions of 8 x 12 classes of speed and direction of wind. Then an average will be taken of the air concentrations obtained ($\chi_{ij}$) with weights proportional to the probability of occurrence of the wind classes ($f_{ij}$).

The resuspension rate $t_r$ [$s^{-1}$] is assumed to depend on the wind velocity (Anspaugh et al 1975) through an expression envolving the friction velocity $v_*$:

$$t_r = C\, v_*^3 \qquad C \approx 10^{-10}\ s^2/m^3 \quad \text{(eq.1)}$$

The friction velocity can be related with the wind speed v at the height z where it has been measured assuming a logarithmic profile of the wind speed with the height:

$$v_* = v\, k \,/\, \ln (z/z_0) \qquad \text{(eq.2)}$$

which is a good aproximation for situations with neutral stability, being k is the Von-Karman constant and $z_0$ is the soil roughness.

The sinks of activity in the volume element are deposition to the ground as well as difusion towards higher layers.

The activity g deposited per unit of surface and time ($Bq/m^2s$) is:

$$g = v_g \chi (x,y) \qquad \text{(eq.3)}$$

where $v_g$ is the dry deposition velocity for the kind of particles considered (m/s), [Sehmel 1984] and $\chi$ is the air concentration ($Bq/m^3$).

Wet deposition is neglected, as it is very small compared with the dry deposition. This is reasonable in arid regions like the studied one.

The activity flowing to higher layers is:

$$f_v = D_z \, \partial\chi/\partial z \qquad \text{(eq.4)}$$

where $D_z$ is a phenomenological coefficient of turbulent diffusion ($m^2/s$) which varies with the atmospheric stability. In our case (eq.4) will be aproximated by:

$$fv = D_z \chi / h \qquad \text{(eq.5)}$$

where h is the height of the compartment (typically between 2 and 2.5 m).

This approach is valid if $\chi(h+h/2) << \chi(h/2)$, i.e. if the mean concentration in the layer of heights [h, 2h] is negligeable in comparison with the mean concentration in the layer that is in contact with the soil.

In relation to the horizontal flow, there are two types of flow transfering activity from a volume element to other: advection due to the wind and difusion due to collisional phenomena and microturbulence in the

air masses. These flows are:

Advective: $F_c = [\delta x \delta y\ h]\ v \cdot \nabla \chi$

Diffusive: $F_d = [\delta x \delta y\ h]\ \nabla(D \cdot \nabla \chi) = [\delta x \delta y\ h]\ D \cdot \nabla^2 \chi$

where:

Fc: is the activity gained through advection by the volume element [δxδy h] per unit of time (Bq/s).

v: the wind velocity (m/s).

$F_d$: the activity losed through diffusion by the volume element per unit of time (Bq/s).

D : The effective diffusion coefficient for the type of stability and size of particles considered ($m^2/s$).

The equation for the balance of activity in the volume element [δxδyh] is (writing the balance per unit of surface):

$$\frac{d}{dt} h \chi = t_r C_s(x,y) - v_g \chi - h\ v \cdot \nabla \chi - h\ D\ \nabla^2 \chi \quad \text{(eq.6)}$$

Two assumptions will be made at this point:

The first one is that the contribution of the transient states to the annualy averaged air concentration is negligeable in comparison to the contribution of the steady states associated to the different classes of wind velocity. Therefore, equation (6) will be solved with the condition d (hχ)/dt = 0.

The second approach is that -h D $\nabla^2 \chi$ is much smaller than the remaining terms in the equation (6) and so its contribution to the

horizontal flow can be neglected.

With these approaches and (Eq.5) the equation (6) becomes:

$$h\, v \cdot \nabla \chi = t_r\, C_s(x,y) - v_{ef}\, \chi \qquad \text{(eq.7)}$$

where $v_{ef} = v_g + D_z/h$ (eq.8)

This problem is straightforward taking a coordinates system in which the wind velocity is parallel to one axis.

The algorithm used has been detailed in (García-Olivares 1992).

The model calculates the annually averaged concentration of activity in each point of a rectangular region containing the contaminated area.

Meteorological conditions in 1988 and air concentrations measured during several years in four points have been used to contrast the model. The model is able to predict the order of magnitud of the air concentration inside a factor of 3 in the points with available measurements.

## 4.4 FURTHER STUDIES

* Simulación studies. Wind tunnel studies.
* To design an to execute more experimental studies.
* To study dry and wet deposition.
* To advance in models. A 3-dimensional advective-diffusive transport of particles will be developed to take into account diffusive effects more accurately.

## 5. FOOD CHAIN

### 5.1 INTRODUCTION

Althought inhalation has been considered the major pathway by wich plutonium reaches man, ingestion of plutonium contaminated foodstuff through the soil-to-plant pathway or by resuspension pathway has been critically evaluated because of the long persistence and general immobility of plutonium in the enviroment.

### 5.2 SOIL TO PLANT TRANSFER

Several studies have been conducted in Palomares to evaluate plant uptake of plutonium under field conditions.(E.Iranzo et all 1988, C.E. Iranzo et all 1989).

Farming procedures in the area are typical of Mediterranean agricultural zones with scarce yearly precipitation in the order of 200 $l/m^2$ requiring artificial means of irrigation. Until recent years irrigation by flooding with water pumped from wells in the area was the prevailing system. At present the drop to drop system is being used.

The main crops in the area are tomatoes, barley and alfalfa. The production of water melons, melons and peppers has largely increased in the last years. Other products represent an small percentage of the total crop.

Plutonium concentrations have been analized in these main agricultural products sampled in six 50 x 50 m study plots along the years since the time of the accident. Vegetation sampling took place within an area of a circunference of a one meter radius, with a center in each of the nine points of each plot where the soil samples were

taken. Each sample correspond to a 5-10 Kg of wet weight. When control of other plots in the zone was stablished in 1978, a random sampling system was implemented taking 5-10 Kg samples of each part of the studied plants.

Our soil-to-plant transfer studies are based in 1850 analisys of the different parts of the main crops, tomato, barley and alfalfa.

Soil periodic sampling has been also conducted from nine points along the diagonals of the six study plots, equidistant from each other. Each soil sample was 30 milimeters diameter and 45 centimeters deep divided into five sections.

Concentration ratios have been calculated using average concentrations values ( E.Iranzo et all 1988 ). The values cooresponding to the main crops are presented in the table 5.1

**Table 5.1 Soil to plant concentration ratios**

| PLANT | | CONCENTRATION RATIO |
|---|---|---|
| SPECIES | PART | |
| Tomatoes | Fruit | $1.5 \times 10^{-4}$ |
| " | Washed fruit | $0.9 \times 10^{-4}$ |
| " | Plant | $2.3 \times 10^{-3}$ |
| Barley | Grain | $1.9 \times 10^{-3}$ |
| " | Straw | $5.0 \times 10^{-3}$ |
| " | Spicule | $6.2 \times 10^{-3}$ |
| Alfalfa | Edible | $8.9 \times 10^{-3}$ |

### 5.2.1 Conclusions

* Plutonium is inhomogeneus distributed in the soil, but due to farming practices over the years, homogeneity is increasing.

* Not all the vegetables samples studied, even from the same plot , present plutonium contamination higher than the minimum detected by our analytical procedures.

* The highest pecenteage of the samples labeled positive correspond to those parts of the plants wich present larger surface or higher posibilities of surface retention of particles.

* Only the 6% of the washed tomatoes samples show plutonium contamination that conduce us to think that the greatest part of the plutonium concentration activity is due to external surface contamination and not to the root uptake.

* In Mediterranean climatological places with scarce precipitations resuspension play an important role. External surface contamination produced by resuspension represent the most important process that produce plutonium contamination of the agricultural products.

* The values of the soil-crop concentration ratios are in the order of $10^{-4}$ for tomato fruit and $10^{-3}$ for the tomato plant and for the diverse components of barley and alfalfa.

* Contamination in the vegetation and the crops of the area persist along the years.

## 5.3 ANIMALS

Plutonium and Americium input to animals is due to ingestion of soil and vegetation and to inhalation. Plutonium could reach man by

ingestion of milk or meat from animals raised in the contaminated area.

Studies has been conducted on Palomares area in order to evalute the plutonium and americium internal contamination of animals potentially constituent of the Palomares people diet.

Like it is well known costumes influence very much the main diet of the villages. In Palomares area still remain alive the usage of pasturing. The main catle are goats, being milk of goats together with milk of cow from the small dairies a usual component in the diet. Another habit is to fatten hens and chickens for family consum, feeding them with products cultivated in the area.

Therefore chickens and hens fatten in this conditions were taking as samples, with the aim to determinate plutonium and americium activity concentrations in their organs and bone. At the same time Pu-239 + Pu-240 and Am-241 concentration in milk has been determined in several samples taken some years from goats grazing in the area for several years and in milk from dairies.

Some animals can be used as an indicator of the enviroment contamination. This is the case of snails, that can be used as indicator of the contamination in the soil.

In order to know activity concentration in snails and how well they reflect the activity concentration of the soil where they live, snails have been sampled in the differents soil study plots. Flesh was separated from shell.

### 5.3.1 Conclusions

* Plutonium values higher than the minimum activity concentration detected by our analytical method has not been determined in the visceras and flesh of the hens and chickens analysed.

* Values of Pu-239+Pu-240 higher than the minimum activity concentration detected by our analytical method determined in the plume chicken samples analized seem to be due to external contamination. This animals have been fatten totaly free, moving in the area.

* The data obtained from the snail samples show a strong relation between plutonium concentration in soil and in the flesh of the snails. Concentration in shell was lower than in flesh.

* The highest value of plutonium concentration activity measured on goat milk is 41.8 $\pm$ 7.38 mBq/l. The ingestion of one liter of milk during one year by the critical group, babys below one year, would represent a Committed effective dose equivalent, in 70 years of 5,5 µSv.

## 5.4 INGESTION DOSE ASSESMENT

Field study data obtained through the observation of crops cultivated in contaminated soils by plutonium has permit us to deduce individual and collective doses derived from the ingestion of these products.

Annual harvest has been deduced, based on the medium of the data obtained yearly regarding the productivity of the crops. The data used for calculation has been the following:

Tomatoes: 80.000 Kg/Ha

Barley :

Grain : 2.500 Kg/Ha

Straw : 2.200 Kg/Ha

Alfalfa : 7000 Kg/Ha

For calculation has been consired that crops has been cutivated

in the plot with a Pu-329+Pu-240 concentration activity of 2.1 KBq x $kg^{-1}$, and the fact that all harvest is contaminated.

table 5.2 shows the values of the collective committed effective dose equivalent, nov Committed effective dose (Iranzo et all 1988). These have been determined after condidering soil-crop concentration ratios determined ( Iranzo et all 1988), the ingestion to milk transfer coeficients specified for terra code (ref. ) and the highest values of Sv/Bq ratios (ref. ) for the estimation of the committed effective dose equivalent by means of the ingestion of Pu-239+Pu-240 by individuals of different ages.

**Table 5.2 Collective committed effective dose equivalent**

| CROP | | | TOMATOES | BARLEY | | ALPHALPHA |
|---|---|---|---|---|---|---|
| | | | | Grain | Straw | |
| $^{239}Pu + ^{240}Pu$ IN ANNUAL CROP Bq x $Hd^{-1}$ x $y^{-1}$ | | | $25.2 \times 10^{3}$ | $9.98 \times 10^{3}$ | $23.1 \times 10^{3}$ | $130.8 \times 10^{3}$ |
| DIRECT CONSUMPTION $S_{E,70}$ (man Sv) | | Adult | $3.02 \times 10^{-3}$ | | | |
| | | 5 y. old | $6.05 \times 10^{-3}$ | | | |
| INDIRECT CONSUMPTION $S_{E,70}$ (man Sv) | Beef | Adult | | $6.0 \times 10^{-10}$ | $1.3 \times 10^{-9}$ | $7.9 \times 10^{-9}$ |
| | | 5y. old | | $12.0 \times 10^{-10}$ | $2.7 \times 10^{-9}$ | $15.7 \times 10^{-9}$ |
| | Milk | Adult | | $1.2 \times 10^{-10}$ | $2.8 \times 10^{-10}$ | $1.6 \times 10^{-9}$ |
| | | 1y. old | | $3.6 \times 10^{-10}$ | $8.32 \times 10^{-10}$ | $4.7 \times 10^{-9}$ |

### 5.4.1 CONCLUSIONS

. The direct Ingestion of non washed tomatoes produce by hectare, would represent as, a maximum, a collective committed effective dose equivalent of $3.0 \times 10^{-3}$ man Sv x $year^{-1}$ for adults.

. The individual dose would be on the order of 1.5 μSv x $year^{-1}$ when based on a yearly consuptiom of 40 Kg of tomatoes.

. Cultivation of products wich are used for feeding domestic animal decreases considerably the value of collective dose for the public. The dose derived from the compsution of meat of animals feed with this products by adults runs between $0.6 \times 10^{-9}$ and $7.9 \times 10^{-9}$ man Sv x $year^{-1}$. The dose derived from the consumption of milk by babies is in the range between $0.4 \times 10^{-9}$ and $4.7 \times 10^{-9}$ man Sv x $year^{-1}$.

## 5.5 FURTHER STUDIES

* To study Am soil to plant transfer factors.
* Autoradiographic studies on vegetables.
* Studies about Hot Particles.
* Studies about transfer factors in animals.

## 6. MARINE ENVIROMENT

In 1985, before Chernobyl accident, a wide research programme was achieved with the purpose of studing the consequence of the accident (1966) in the marine ecosystem, selecting sediments as a tool to measurement radionuclides depositional history in the period from 1966 to 1985.

Other objectives were to determine the distribution of transuranics and other long-lived radionuclides in the marine sediments of three depth marine regimenes, comparing this area with an area not affected by this accident.

(This project was partially funded by CEC in Radiation Protection Programmes)

To carry out these objectives were done:

- five samplings, on board of the vessels Coornide Saavedra and Jafuda Cresques, picking out sediments from 34 stations between Cape Palos and Gata (Fig.1) and determining the seasonal particulated matter variation .

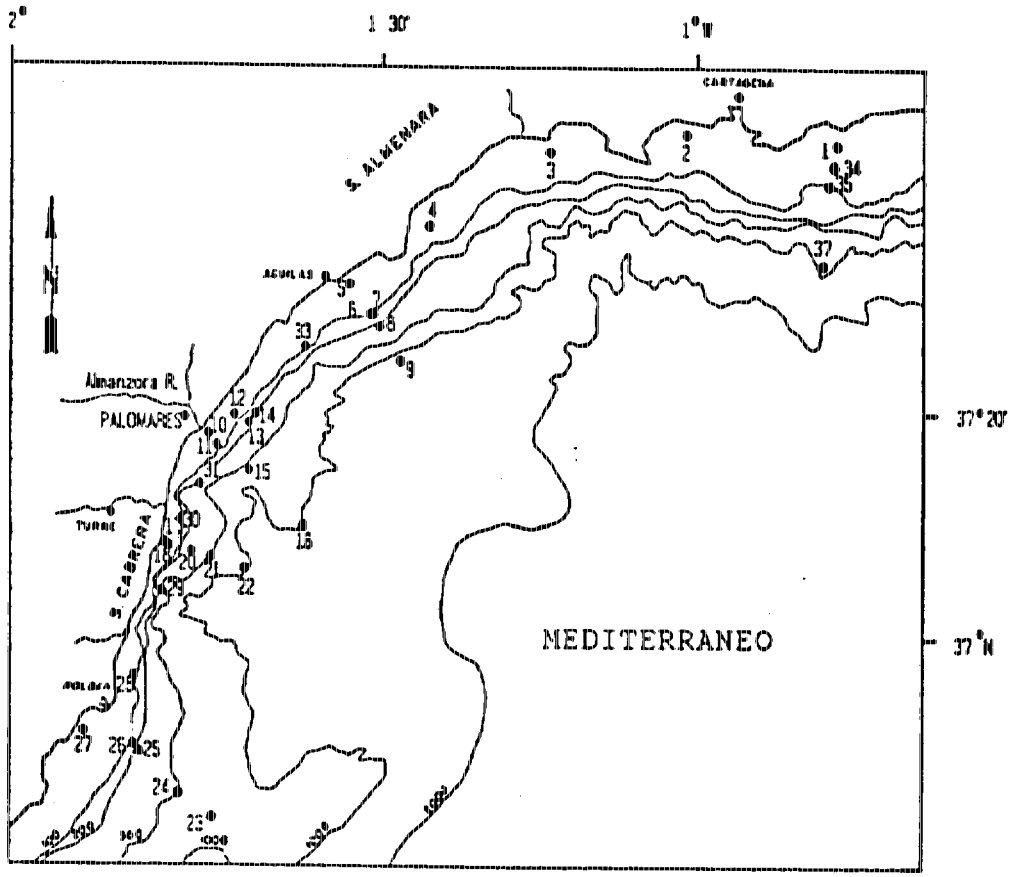



Figure 1 : Sampling Stations

- Analysis of $^{239+240}Pu$, $^{241}Am$ , $^{137}Cs$, $^{210}Pb$ in sediment cores and surface sediments
- Geochemical and grain size composition

The main conclusions are published in the literature[1,2,3,4,5,6] remarking that :

-The radionuclides concentration decreases as water depth increases (Fig.2) and also increases at the south of Almanzora river mouth, decreasing along the coast (Table 1)

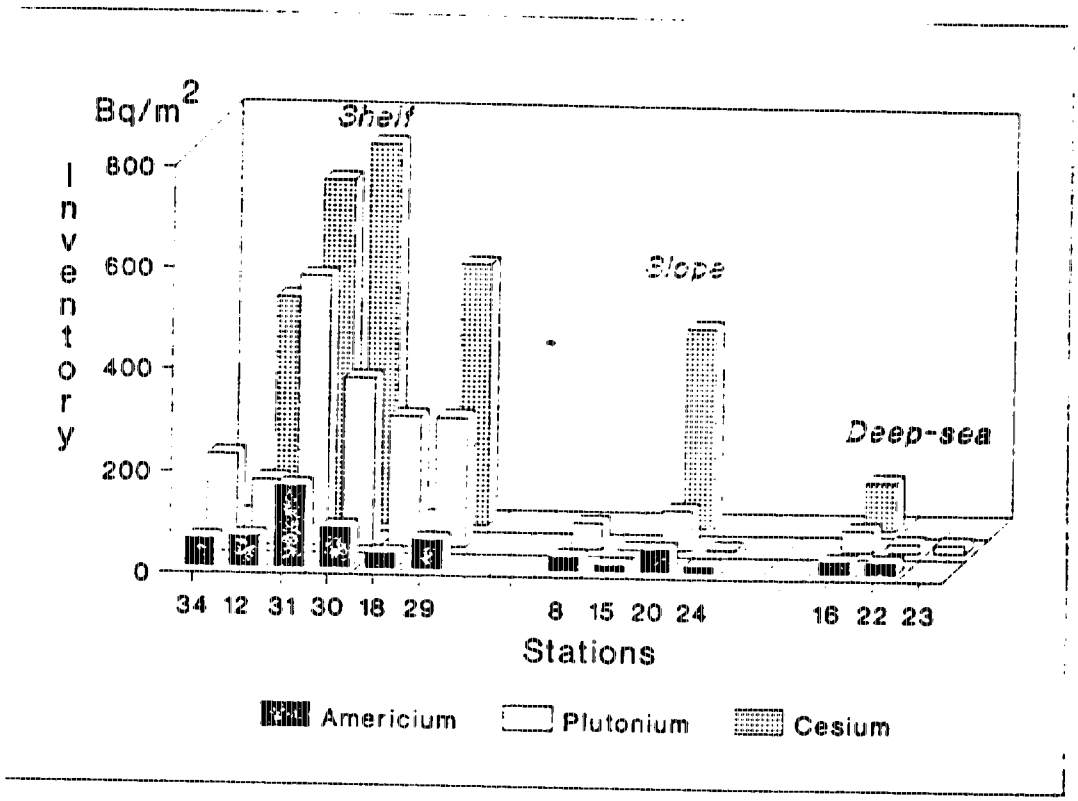



Figure 2 : Inventories in three depth regimenes

-The radionuclides concentrations are similar to those found in others Mediterranean areas except in one small area located south of the Almanzora river mouth.

-The importance of transport the radionuclides by floods in Mediterranean areas with a very low rainfall index .

Table 1

Contents of $^{239+240}Pu$ and $^{241}Am$ in surface sediment. Concentration expressed in Bq/Kg dry weight ($\pm 1\sigma$)

| Station | $^{239+240}Pu$ Activity $\pm 1\sigma$ | $^{241}Am$ Activity $\pm 1\sigma$ |
|---|---|---|
| Continental Shelf | | |
| 01 | 1.55 ± 0.15 | ---------- |
| 02 | 1.17 ± 0.10 | 0.49 ± 0.09 |
| 03 | 0.92 ± 0.08 | ---------- |
| 05 | 1.12 ± 0.07 | ---------- |
| 06 | 0.77 ± 0.07 | ---------- |
| 07 | 0.18 ± 0.03 | ---------- |
| 10 | 0.43 ± 0.07 | 0.71 ± 0.20 |
| 11 | 5.22 ± 0.32 | 1.95 ± 0.40 |
| 12 | 1.80 ± 0.13 | ---------- |
| 13 | 0.48 ± 0.07 | ---------- |
| 17 | 0.38 ± 0.05 | ---------- |
| 18 | 1.72 ± 0.17 | ---------- |
| 20 | 0.73 ± 0.10 | 0.55 ± 0.10 |
| 25 | 0.72 ± 0.07 | ---------- |
| 26 | 1.15 ± 0.08 | 0.76 ± 0.09 |
| 27 | 1.32 ± 0.08 | ---------- |
| 28 | 1.92 ± 0.17 | 0.93 ± 0.10 |
| 29 | 1.93 ± 0.12 | 0.39 ± 0.07 |
| 30 | 2.47 ± 0.20 | ---------- |
| 31 | 2.43 ± 0.18 | 2.94 ± 0.28 |
| 33 | 0.67 ± 0.08 | ---------- |
| 35 | 1.05 ± 0.10 | ---------- |
| Slope and deep-sea | | |
| 16 | 0.43 ± 0.05 | ---------- |
| 22 | 0.09 ± 0.02 | ---------- |
| 09 | 0.69 ± 0.08 | ---------- |
| 21 | 1.09 ± 0.07 | ---------- |

- $^{241}Am$ (Not calculated)

In the period from 1990 until now a new project was started, focussing the attention on the Continental Shelf off Palomares (Fig.3). This sensitive Environment is considered an area of economical interest due to the fisheries activities. Our participation was included in a multinational Project in which three European Institutions were involved making the study of the area possible and comparing it with other marine ecosystems. (Institution/area of interest)

ENEA ( Ligurian Gulf), CEA La Hague Estuary (France), UCD Irish sea (Ireland), CIEMAT ( Palomares)

The main objectives are:

-To study the radionuclides distribution in Almanzora river gulch and beach edge

-To determine the role of submarine canyons in the transference of radionuclides from Continental shelf to deep sea areas (Palomares Canyon) and

- To study the influence of terrigenous contribution in the increase of radionculides concentration in the Continental Shelves.(Palomares and Mediterranean areas)

The data are unpublished until now, but will be published to due time.

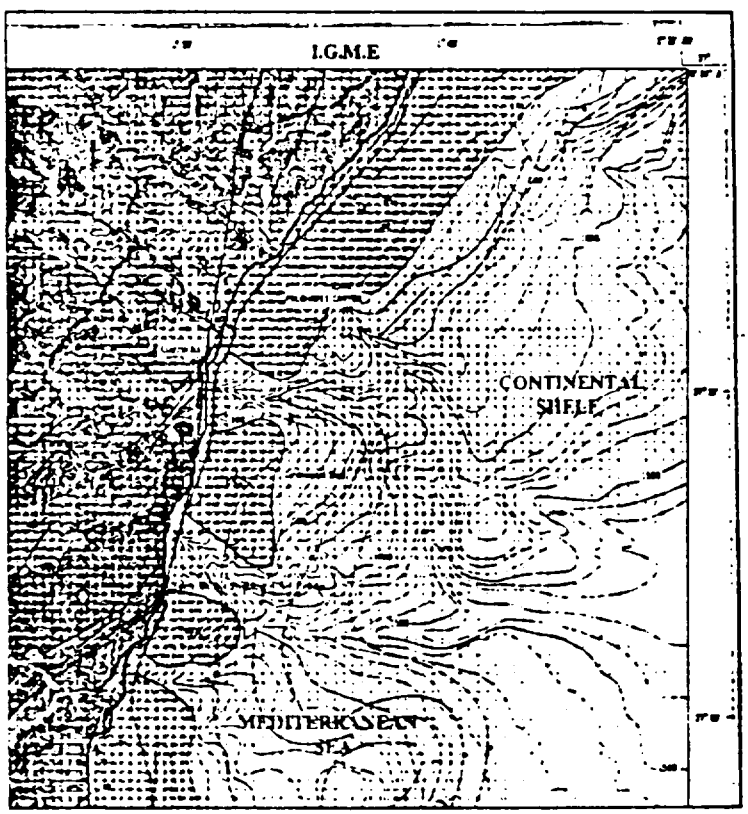



Figure 3 : Palomares Canyon. Objective of the next studies.

# 7. DOSES ASSESSMENT

## 7.1 INTRODUCTION

Assessment internal contamination caused by taking in Pu, whatever the way this might have occurred (ingestion through contaminated food, inhalation and depositing on the skin), is based mainly on determining the concentration of Pu in excreta (indirect measurements).

Animal experiments performed in order to determine absortion through the gastro-intestinal tract have shown very low values. This fact, together with the low percentage of absortion of the element by plant roots, leads us to regard this form of contamination for people living in the Palomares zone as being negligible.

Absortion through the skin cannot be a risk for the inhabitants of an area contaminated by Pu since the skin is an efficient protection barrier and should only be taken into consideration in the event of being attacked by aggressive agents.

Inhalation of plutonium, mainly in the form of oxides, is the greatest internal contamination risk for the population being studied. A mathematical model developed at the ICRP expresses the deposits in the various lung compartments according to the grading of the particles expressed in terms of aerodynamic size. The spacial distribution of the particles in the lungs, as well as the granulometric study of the soil and the physical-chemical composition of the latter, linked to the existing activity, is an important parameter when it comes to studying the risk attached to the volume of lung tissue affected by radiation.

Elimination of the particles deposited in the upper air passages (greater than 10 $\mu$) is very rapid, seldom taking longer than two days. Elimination through the gastrointestinal tract by means of deglution will probably be the important source of activity for the faeces.

Particles of under 10 μ, being those which remain in the different areas of the lung, are eliminated, after remaining there for various lengths of time, by means of two different mechanisms, namely macrophages and by passing through the alveolar-capillary barrier. The chemical form that conditions the hydrolysis and complexation mechanisms plays an important role in the lung cleansing kinetics. The fraction deposited in the pulmonary region is that formed by particles smaller than 10 μ. Studies carried out in soil-sampling, performed in November 1986 in an area close to the impact point of one of the fractionated nuclear bombs (E. Iranzo et al. 1990), brought to light an abundance of 20.1 per cent for this fraction, with an average activity of 919 Bq/g.

The plutonium fraction entering the different organs and tissues from the concentration present in the blood depends mainly on the stability of the complexes to be found within the latter.

Plutonium is eliminated via the urine and the faeces. Since elimination via the urine is more representative of the activity in the bloodstream, this form of excretion was chosen for our tests, using 24-hour urine (having corrected the volume by means of a parallel creatinine analyses).

## 7.2 MEASURING INTERNAL CONTAMINATION

Internal contamination is determined by means of direct anthropogammametric measurements on a body counter, and by means of indirect measurements in excreta.

All people over the age of thirteen living in the Palomares zone have been analysed at least once by means of direct measurements, and all these measurements have given values below the Detection Limit. The number of measurements in whole body counter is 1190 for 769 persons. In addition they have been analysed at least once by means of indirect bio-elimination measurements in 24 hour urine samples. Only 8 per cent of the analyses carried out have given results higher than the Minimum Activity Detected

by our analytical method (MDA).

The total number of analyses performed up to 1990, as well as the number of times each person was analysed, can be seen in table nº 7.1.

The Pu bio-elimination studies since 1985 are complemented with sequential Am anlyses. These Am tests had not been performed previously due to the fact that this element comes from Pu 241 which has an average life of 13 years. It is significant to point out that no one has given results higher than the MDA more than once, and it is also important to stress that, except on rare occasions, positive results for Am had previously been positive for Pu, with these cases being in fact women aged around forty-five.

## 7.3 DOSE CALCULATION

The criteria and techniques used for estimating the committed effective dose equivalent in the case of those persons considered as being internally contaminated by plutonium on account of the results of the excretion of $^{239}Pu$ + $^{240}Pu$ in the urine, have been as follows:

The intake is due to acute inhalation of particles of 1 μm (AMAD), class Y plutonium at the time of the accident or during the days inmediately afterwards, for the people who were in Palomares when it happened.

Chronic inhalation was rejected upon discovering that the positive values of excretion in urine that were obtained would require the concentration of plutonium activity in the surrounding air to be constantly very much higher than the plutonium-concentration values measured in the area. The average concentration in air measured in the urban zone was around 5 $\mu Bq/m^3$ and about ten times higher in the area with the highest concentration level.

In order to fix the date of the probable intake due to inhalation in the case of those people who were not in the area or who had not been born

in 1966, account has been taken of the data on which they came into the area, the type of work and habitat, the years in which, owing to particular circunstances, higher concentration of Pu in the air were measured in the zone, and the correlation of the Pu values in urine with the excretion curve.

Determining the activity of Plutonium intake through inhalation on the basis of the values of excretion in urine on the date of the sample, by means of the method developed by K. Eckerman (Ec 84) in the Oak Ridge National Laboratory. This method is based upon the metabolic intake and pulmonary distribution model of the ICRP and on the urinary excretion model of Langham modified by W. Moss (Moss 83) in Los Alamos National Laboratory. In the case of those people who have had Pu measurements in urine higher than the MDA, it was considered that their MDA values were positive and with a value equal to the limit.

Calculation of the committed effective dose equivalent by means of the Sv/Bq conversion factors recommended by the ICRP publication 30 in order to calculate the weighted committed dose equivalent for the most important organs from the point of view of their contribution to the committed effective dose equivalent.

For experimental purposes, estimations have also been made of the committed effective dose equivalent for 70 years, using for persons of different ages the Sv/Bq conversion factors established by Kaul and his colaborators (Henrichs K. et all, 1985).

The results of the measurements of $^{239}Pu$ and $^{240}Pu$ in the urine samples taken over a period of more than 20 years from those Palomares residents who have been checked at least once, as well as the estimations of the committed effective dose equivalents which, according to the results, have been considered as being contaminated, are discussed below. Pu in urine above MDA was measured in 57 people.

The estimation of the committed effective dose equivalent was carried out on the 55 people who were considered really to have suffered internal

contamination, by making a rigorous study, for each of them, of the Pu values in urine, the possibility of external contamination of the sample and its greater or lesser correlation with the model developed by K.Echerman on the basis of ICRP pulmonary model and W.Langham's excretion curve modified by W. Moss.

Consequently, by following this procedure, calculations were made for the intake of Pu due to acute inhalation for the 45 people, 24 men and 21 women,who were in Palomares when the accident occurred. In the case of the 10 people, 5 men and 5 women, who were not in the area when the accident took place, it is difficult due to the shape of the curve for the excretion of Pu in urine, to determine the date of intake on the basis of the factors indicated in the methodology. In fact, the doses were estimated in terms of the date of intake considered to be the most likely.

The values of the committed effective dose equivalent estimated for the people in question fall between 20 and 200 mSv (2-20 rem). Table nº 7.2

The study made into the doses calculated for these 55 people reveals a normal logarithmic correlation. Having calculated Henry's straight line, and with the established logarithmic types having been represented, it can be seen that all of them are within the 95% confidence determined by the calculation of Leveau's abacus (Fig.nº 7.1). This fact confirms that the criterion followed for calculating the possible intake by using the averages of the intakes calculated for each excretion is right enough.

In order to study the correlation between the Pu values in urine and Echerman's model, a far- reaching study was made on the 30 people who had shown values higher than the MDA in three or more samples. All of them had been given at least 5 tests during the 22 years following the accident, and 60% of them had a minimum of 9. The adjustment of the curve relating them with the acute inhalation intake on a particular date, led us to the conclusion that, by calculating the intake starting from each one of the values of Pu in urine for the date of the sample, and determining the average value of the intakes that were calculated, we could obtain the most suitable estimation of the intake due to acute inhalation in the case of

each person.

## 7.4 DOSES ASSESSMENT BASED ON AIR CONCENTRATION DATA

Determination of the inhalation risk for people living and farming in the area, and as a consequence, the internal radiation risk for permanent inhabitants, including children, have been carry out in 1985 (E. Iranzo et all. Health Physics 1987.)

This calculation have been done based in a cronic intake, the dose calculation from excretion value consider acute intake. The potential committed dose equivalent for organs during the period 1966-1990 for adults who lived in the urban zone and the people in the cultivated zone with the greatest potential risk, station 2-2 and a mean aerosol size (AMAD) of 1 μm are shown in tables 7.3 and 7.4.

The contribution of the committed dose equivalents in the five organs to the potential committed effective dose equivalent to people during the 24-y period 1966-1990 is 0.2123 mSv (21 mrem) in the urban area and 1.9653 mSv (196 mrem) in the area which corresponds to the critical group.

## 7.5 OTHER ACTIVITIES RELATED TO THIS AREA

CIEMAT has recently been participating in an intercomparison exercise involving nine European laboratories, organized by EURADOS, for the purpose of calculating the doses received by five workers contaminated with different radionuclides, including different Pu and Am isotopes. This intercomparison has recently been published. (J.A.B. Gibson et all 1992).

Likewise, and as front-line research aimed at setting up transfer parameters in lungs for dust contaminated with plutonium and americium as a result of an accident, CIEMAT is collaborating with NRPB in a proposal put before the EEC, the title of which is THE INTERNAL DOSIMETRY OF PLUTONIUM AND AMERICIUM RESULTING FROM THE INHALATION AND INGESTION OF DUST PRESENTED

IN AN ACCIDENTALLY CONTAMINATED ENVIRONMENT. The work currently being performed in this field is:

- To provide an experimental basis for assesing the internal dose to humans resulting from intakes of plutonium and americium bearing dusts present in a defined accidentally contaminated enviroment. Part of the work will involve biokinetic studies of these actinides in rats after deposition of dusts in the lungs and gastrointestinal tract.

_ Characterization of the contaminated dust with respect to particle size, chemical and mineralogical composition, and correlation of these characteristics with the Pu and Am concentrations and the physico-chemical forms of these actinides. An important aspect of the study will be the correlation between the transportability of the actinides in-vivo and the results of measurements conducted on the mineralogycal composition of the dusts and the physico-chemical forms of the actinides. This could provide a rational basis for assesing the radiological impact of inhaling or ingesting contaminated dusts present in other accidentally contaminated environments.

- We will undertake short-term in vitro dissolution experiments to asses the range of solubility of Pu and Am present in various dusts fractions. The in vivo experiments carried out at NRPB will reflect this range. CIEMAT will also undertake long-term dissolution experiments on the dusts used for the in-vivo experiments in order to compare the dissolution rates observed in-vitro and in-vivo.

- Dose factors per unit intake for this dust will be calculate.

7.6 CONCLUSIONS.

The study about surveillance program in the 769 people tested during a period of 25 years, has permit us to attain the following conclusions:

- As a result of the accident, part of the population, suffered internal contamination due to inhalation.

- It seems to follow that a group of 10 people may have been internally contaminated on account of their habitat and their participation in the agricultural activities of the area, without it really being possible to establish the time in which the intake could have taken place.

- The 70 years committed effective dose equivalent ($S_{E,70}$) for 10 people less than 15 years old, who were internally contaminated at the time of the accident, is only higher then 200 mSv (20 rem) for 1 person (242 mSv). The $S_{E,70}$ for the others 9 people are in the range of 49-157 mSv.

- The assumption made by Legget (Legget, R.W 1985) about a periodical removilization of Pu deposited initially on bone surfaces, could be confirmed in some persons ( 1 man and different women). An increase of Pu urine excretion have been measured about 20 years after in all the cases.

- The Media log normal of committed effective dose equivalent ($S_E$,50) is 92,7 mSv, with the Mediana $\gamma$=79,43 mSv, and varianza $\sigma^2$ =0,31

## 7.7 FUTURE ACTIVITIES

* To continue with the periodic control of the people who live in the area, in order to control their probable contamination, as well as the absence of effects due to radiations.

* It is very important to carry out a comprensive study of those children whose results are higher than AMD.

* To review continuously the hypothesis stablished for the dose assessment, taking into account different hypothesis including cronic intake and different inhalation dates based on the excretion data, habits, etc. Plutonium concentration values have been determined in children that were not borne at the accident and in persons that were not in Palomares at this time. So more hypothesis are being studied.

* Doses assessment taking into account the lung transfer parameters and the doses per unit intake determined by means of the joint NRPB-CIEMAT experiments.

TABLE nº 7.1

TOTAL ANALYSES AND NUMBER BY PERSON FOR THE PERIOD 1966-1989 IN PALOMARES PEOPLE

| nºanalises/ person | Men | Women | Men + Women | nºanalises |
|---|---|---|---|---|
| 1 | 112 | 93 | 205 | 205 |
| 2 | 163 | 160 | 323 | 646 |
| 3 | 37 | 42 | 79 | 237 |
| 4 | 20 | 32 | 52 | 208 |
| 5 | 14 | 13 | 27 | 135 |
| 6 | 14 | 9 | 23 | 138 |
| 7 | 7 | 5 | 12 | 84 |
| 8 | 5 | 6 | 11 | 88 |
| 9 | 9 | 4 | 13 | 117 |
| 10 | 5 | 4 | 9 | 90 |
| 11 | 2 | 6 | 8 | 88 |
| 12 | 2 | 2 | 4 | 48 |
| 13 | 1 | 0 | 1 | 13 |
| 14 | 1 | 1 | 2 | 28 |
| total | 392men | 377women | 769person | 2125analises |

TABLE № 7.2

ESTIMATED COMMITTED EFFECTIVE DOSE EQUIVALENT BASED ON URINARY EXCRETION DATA AND ASSUMED ACUTE INHALATION

| ESTIMATED DOSE mSv (rem) | NUMBER OF PEOPLE |
|---|---|
| < 20 (<2) | 659 |
| 20-50 (2-5) | 22 |
| 50-100 (5-10) | 22 |
| 100-150 (10-15) | 6 |
| 150-200 (15-20) | 5 |

TABLE 7.3

TOTAL POTENTIAL COMMITTED DOSE EQUIVALENT FOR ORGANS DURING THE PERIOD 1966-1990

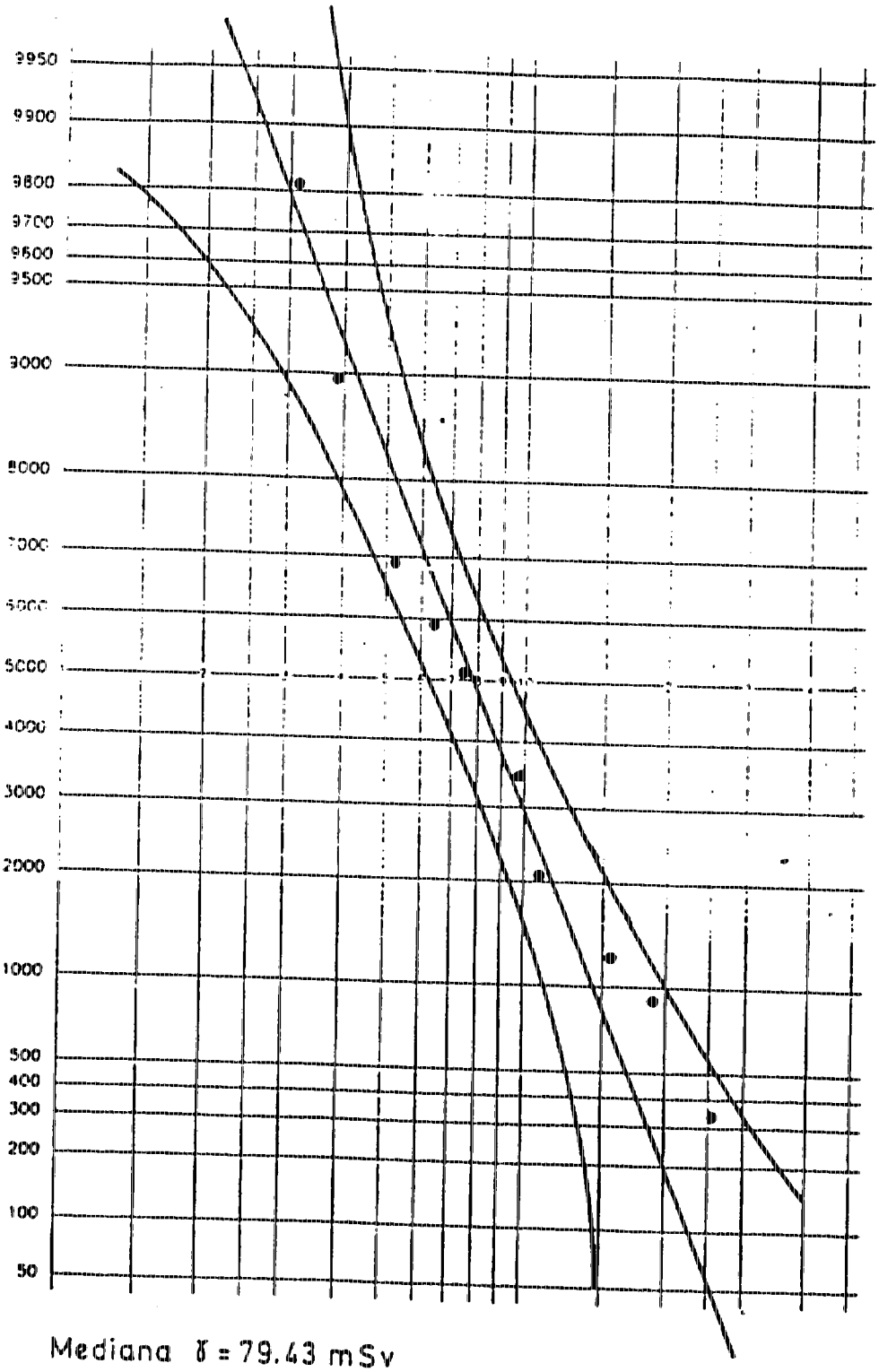
| ORGAN | TOTAL COMMITTED DOSE EQUIVALENT, mSv | |
|---|---|---|
| | URBAN ZONE (P) | STATION 2-2 |
| BONE SURFACE | 1.1243 | 10.5422 |
| LUNG | 0.3987 | 3.5512 |
| LIVER | 0.2520 | 2.3302 |
| RED BONE MARROW | 0.0909 | 0.8435 |
| GONADS | 0.0144 | 0.1331 |

TABLE 7.4

CONTRIBUTION OF COMMITTED DOSE EQUIVALENTS IN ORGANS TO THE TOTAL POTENTIAL COMMITTED EFFECTIVE DOSE EQUIVALENT DURING THE PERIOD 1966-1990

| ORGAN | CONTRIBUTION TO COMMITTED EFFECTIVE DOSE, mSv | |
|---|---|---|
| | URBAN ZONE (P) | STATION 2-2 |
| BONE SURFACE | 0.1349 | 1.2650 |
| LUNG | 0.0478 | 0.4261 |
| LIVER | 0.0151 | 0.1398 |
| RED BONE MARROW | 0.0109 | 0.1012 |
| GONADS | 0.0036 | 0.0332 |

Figure 7.1 DISTRIBUTION OF ESTIMATED $S_{E.50}$ FOR PALOMARES PEOPLE




Mediana $\delta = 79.43$ mSv

Varianza $\sigma^2 = 0.31$

Media log. normal = 92.7 mSv

## ANEX 1. METHODOLOGIES FOR ANALYSIS AND MEASUREMENTS

### 1. INTRODUCTION

An important step in almost all the experimental activities described in the precedents chapters is a chemical separation and measurement of Pu and Am radionuclides . In the most of the cases a ressult in terms of radioactivity concentration must be obtained in order to interpret radiological processes or situations and/or to do further dose calculations. A solid knowledge and experience in radioanalytical work and radioactivity measurement techniques it is therefore essential.

Concerning transuranics, the Enviromental Institute of the CIEMAT hols a very specialised group having working on Indalo Project since many years. Laboratories and measurements facilities enable a high radioanalytical capability.

Addendum I shows a list of specific procedures that are published according to CIEMAT quality control norms.

A short description of alpha spectometry and low energy gamma spectrometry is given below.

### 2. RADIOCHEMICAL ANALYSIS

This techniques require, in general, the following steps:

A) Sample preparation by dry and/or wet separation

B) Separation of interferent elements by precipitatation and/or ionic interchange resins.

C) Purification by means of electrodeposition.

E) Measurement.

The plutonium and americium activity, electrodeposited on the planchet is measured in alpha spectrometry equipment with silicon-barrier semiconductor detectors and a counting time on not less than 1440 minutes.

The lower detection limit, following the Currie Criterion in accordance with the American ASTM Standars (Volume 12,01,1983, c-100) for determining Pu-239 + Pu-240 and Pu-238, with a confidence level of 95%, is :

D.L = 0.3 mBq/planchet

## 3. ALPHA SPECTROMETRY

### 3.1 Description of the equipment

The equipment is made up of two independent modules, model 576-VM ORTEC, with eight 576A double cells, wich means that, in all, there are 32 silicon-barrier semiconductor detectors Mod TR-21-300-100, with a nominal active area of 300 $mm^2$ and a resolution of 21 KeV. Operation voltage is 100 volts and dead time is 0.5 μseconds. The pulses are stored in a ORTEC multichaner analizer with 16368 channels and are processed with the MAEATRO II.

Each 576-VM module is provided with an Edwards high-vaccum pump. Electricity is obtained from a set of storage batteries (UPS) so as to avoid the anomalies caused by transients or pure-duration cuts in the supply from the mains.

### 3.2 Calibration

The detectors are calibrated with secondary Pu-238, Pu-239, Pu-242, Am-241 and Am-243 standars, which are standardized in a proportional gas counter, using the N.B.S. primary standars. The secondary standars used are

from Los Alamos National Laboratory (USA) with their corresponding calibration certificates.

This calibration operation is repeated every six months and the detector backgrounds are determined on a fortnightly basis with a minimum counting time of 1440 minutes.

Plutonium and Americium standars, provided by the N.T.I.S. (U.S.A) are used to check the methods and confirm the worth of the results obtained.

1023 channels are assigned to each detector in order to cover the energy range from 4 to 6 MeV., wich takes in all the isotopes and alpha emitters that concern us.

## 4. AUTOMATIC LOW ENERGY GAMMA SPECTROMETRY SYSTEM (LEPS).

This is made up of two CAMBERRA intrinsic germanium detectors, GL1015 model, one of wich has an inverted cryostat in order to improve the overall efficiency of the measurement, by adding the spectra obtained, in each of the detectors, in a certyain time.

Resolutiom is from 310 eV to 5.9 KeV and from 575 eV to 122 KeV. the 2001 model preamplifier has a counting capacity of up to 100.000 counts per second.

The multichanner analyzer with 4096 channels, 2048 for each detector, is the 3052 model,35 PLUS series made by CANBERRA, connected to a PC by means of a 3575 model interface. This analyzer is extremely versatile, being able to operate in a normal manner or carry out automatic programming sequences.

The calculation of the sum of the spectra, calibration for different geometries, automatic search for peaks, minimum detectable activity, calculation of areas, etc, are achieved by means of the CANBERRA Gamma Spectra Analysis Program, ESPECTRAN AT model, and the 286-model adquisition

and treatment system by the same firm, with graphic output of the spectra on the screen, remote control multichannel handling and control of the sample changer.

The standars used for calibrating the system are from the AMERSHAN firm, with which planchets from the master corresponding to the samples analyzed are prepared.

## ADDENDUM I

## METODOS ANALITICOS PARA LA DETERMINACION DE PLUTONIO Y AMERICIO

1. Procedimiento para la determinación de Pu-239,en muestras de orina,por espectrometría alfa.

2. Determinación secuencial de Am-241,en muestras de orina de 24 horas,por espectrometría alfa.

3. Procedimiento para la determinación de Plutonio-239 + Plutonio-240 en muestras de vegetación.

4. Procedimiento para la determinación de Plutonio-239 + Plutonio-240 en tejidos y muestras biologicas.

5. Procedimiento para la determinación de Plutonio-239, 240 en muestras de suelos.

6. Procedimiento para la determinación de Plutonio-239, 240 y Americio-241 en muestras de aire.

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